Quantum embedding in electronic structure theory

Part 3: Exact embedding of localised orbitals for non-interacting electrons and extension to strongly correlated electrons

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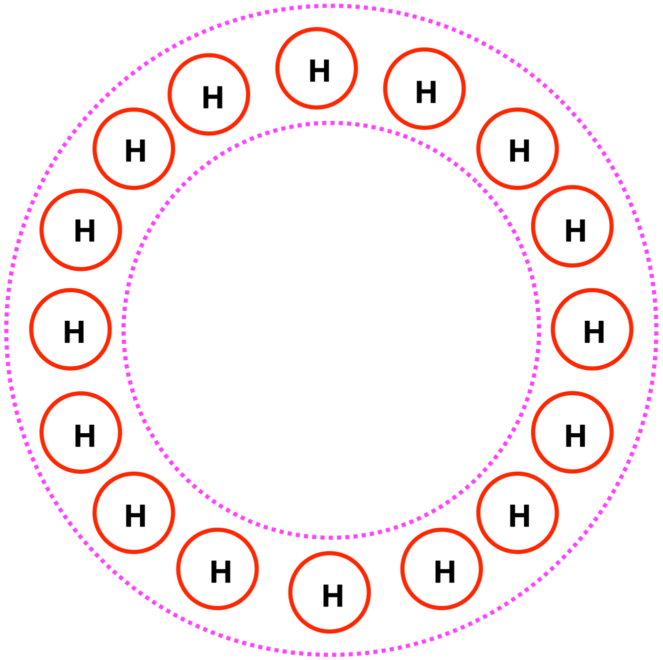
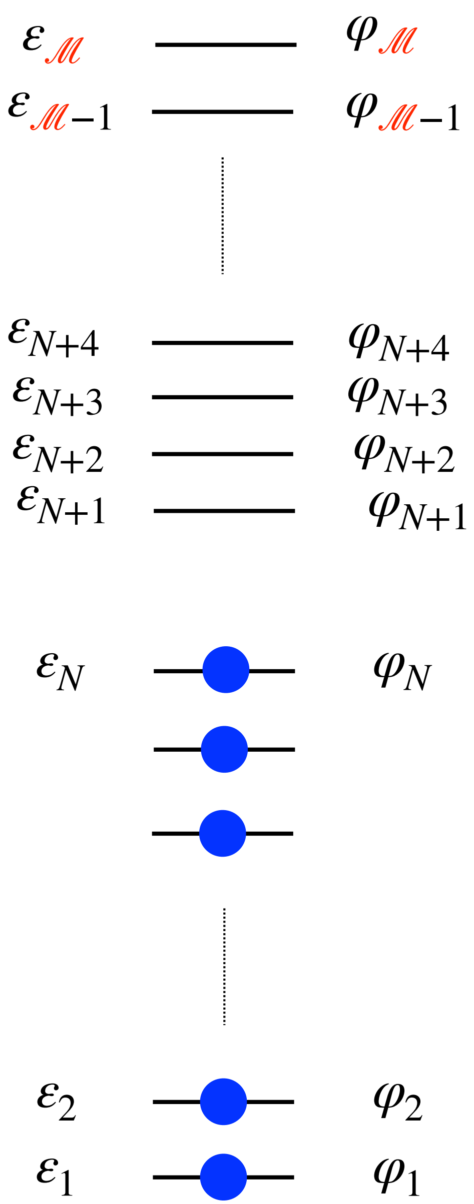
Non-interacting delocalised representation



$$\hat{H} \equiv \sum_{PQ} \langle \varphi_P | \hat{h} | \varphi_Q \rangle \, \hat{a}_P^\dagger \hat{a}_Q$$

Non-interacting (delocalised) molecular orbital representation

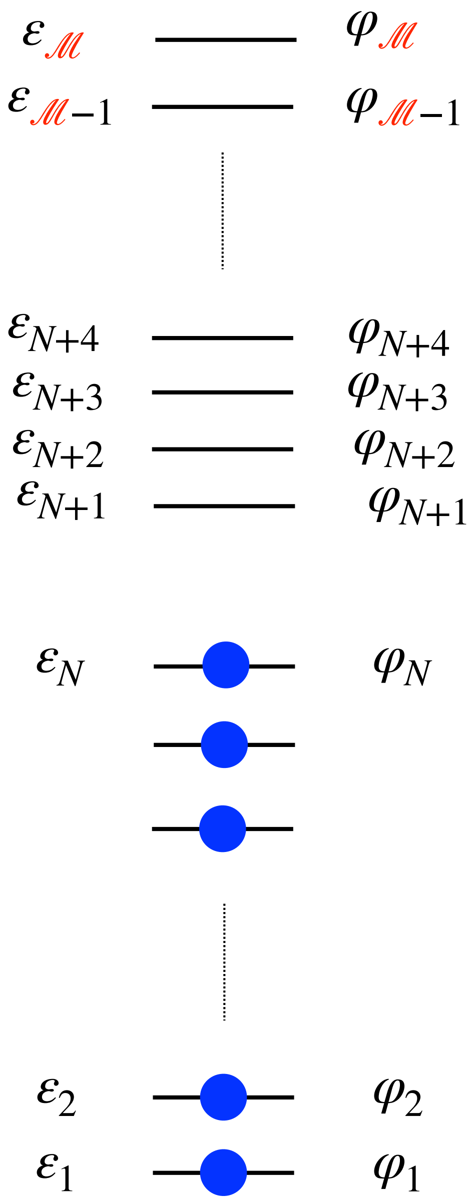


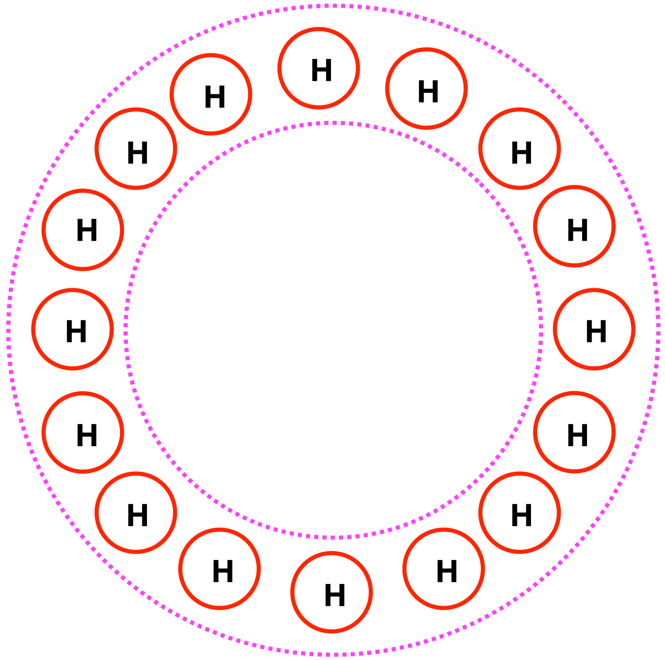
$$\hat{H} \equiv \sum_{PQ} \langle \varphi_P | \hat{h} | \varphi_Q \rangle \, \hat{a}_P^\dagger \hat{a}_Q$$

The molecular spin-orbitals are simply obtained by solving the **one-electron Schrödinger equation**

$$\hat{h}\varphi_Q(\mathbf{x}) = \varepsilon_Q \, \varphi_Q(\mathbf{x})$$

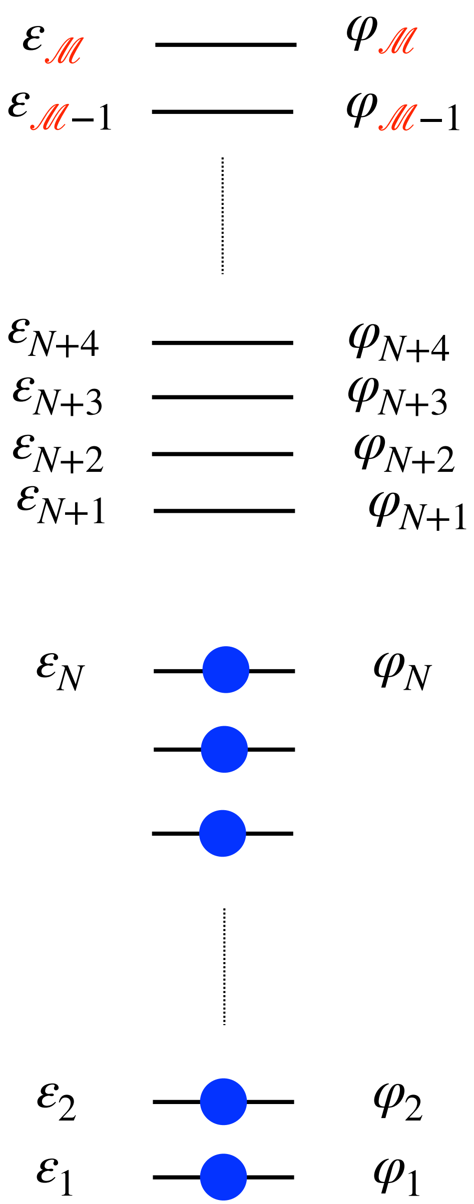
Non-interacting (delocalised) molecular orbital representation





$$\varphi_P(\mathbf{x}) = \sum_{\nu} C_{\nu P}\, \chi_\nu(\mathbf{x})$$

Non-interacting (delocalised) molecular orbital representation



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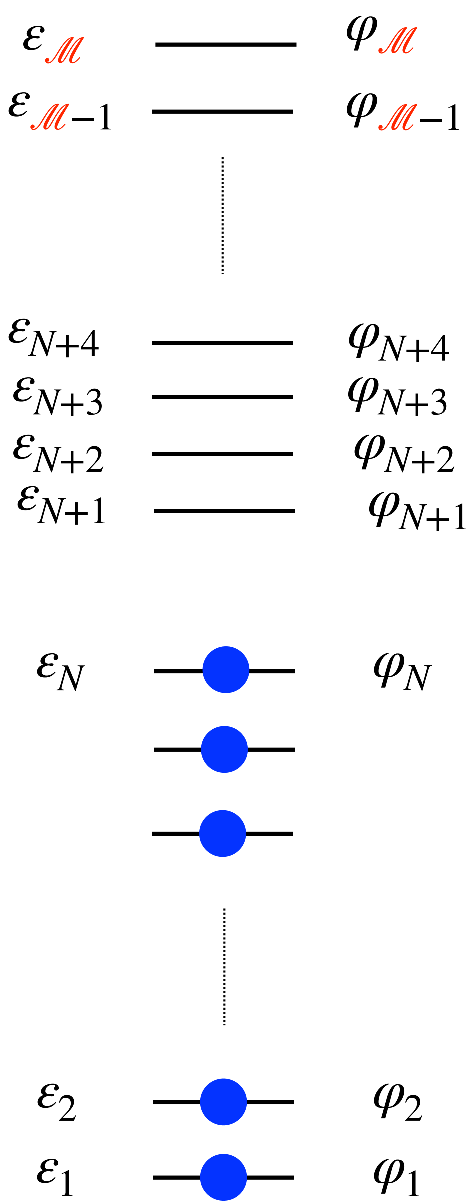
$$\langle \varphi_P | \hat{h} | \varphi_Q \rangle = \varepsilon_P \, \delta_{PQ}$$

Non-interacting (delocalised) molecular orbital representation



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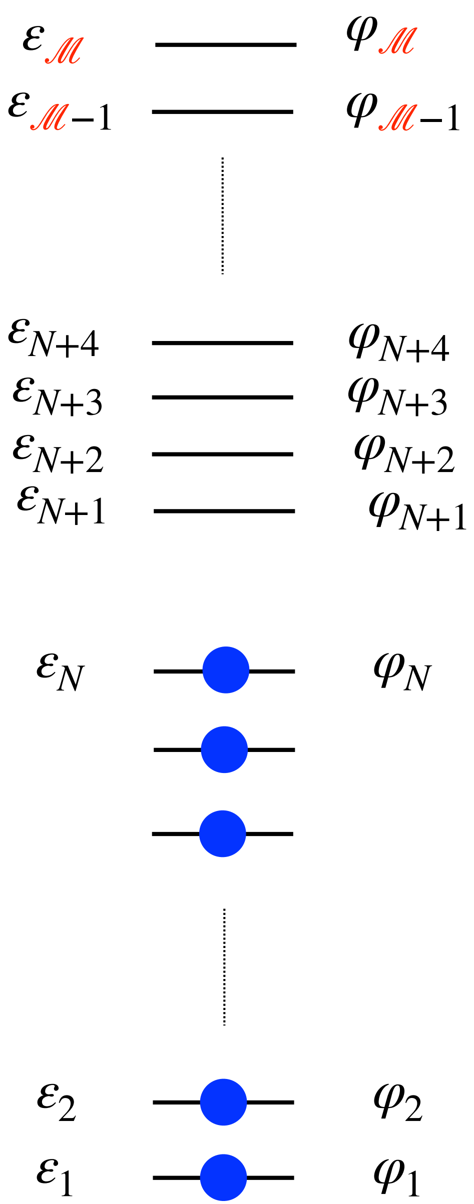
Non-interacting (delocalised) molecular orbital representation

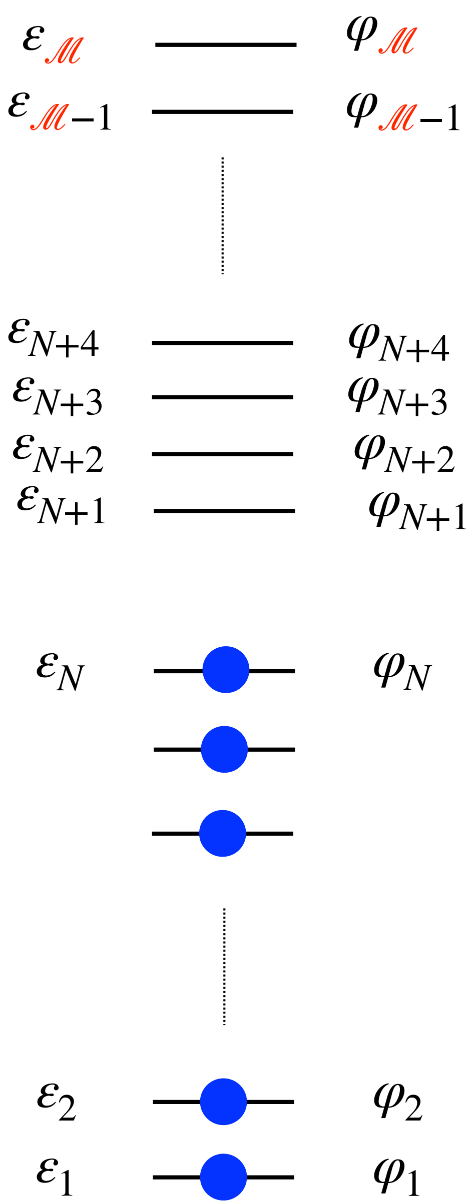


$$\hat{H} \equiv \sum_{PQ} \langle \varphi_P | \hat{h} | \varphi_Q \rangle \, \hat{a}^\dagger_P \hat{a}_Q$$

$$= \sum_{P} \varepsilon_P \, \hat{a}^\dagger_P \hat{a}_P$$

The exact solutions to the non-interacting Schrödinger equation are Slater determinants $\hat{a}^\dagger_{P_1} \hat{a}^\dagger_{P_2} \ldots \hat{a}^\dagger_{P_{N-1}} \hat{a}^\dagger_{P_N} | \mathrm{vac} \rangle$

Non-interacting (delocalised) molecular orbital representation



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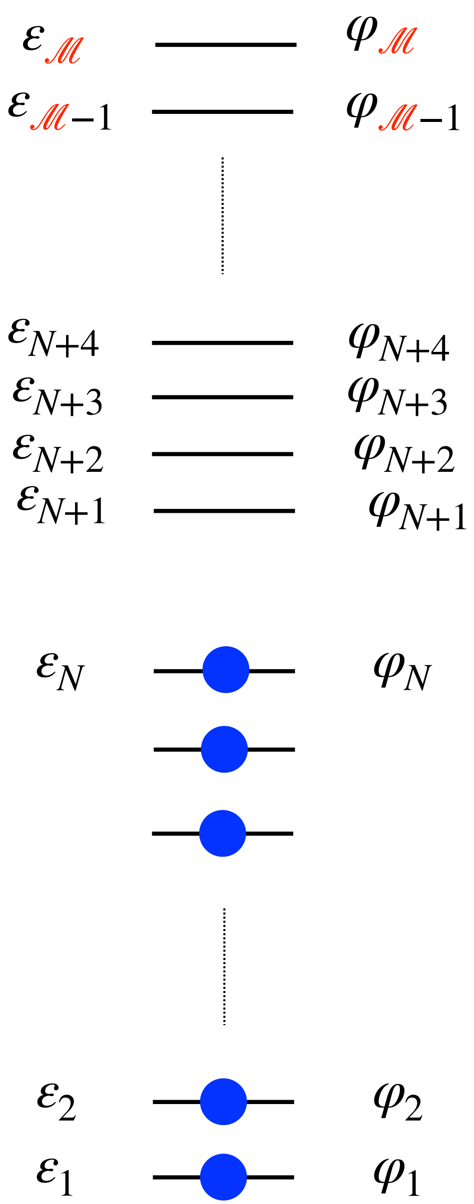
$$\hat{H} \, \hat{a}_{P_1}^\dagger \hat{a}_{P_2}^\dagger \ldots \hat{a}_{P_{N-1}}^\dagger \hat{a}_{P_N}^\dagger | \text{vac} \rangle = \left(\sum_{i=1}^{N} \varepsilon_{P_i} \right) \hat{a}_{P_1}^\dagger \hat{a}_{P_2}^\dagger \ldots \hat{a}_{P_{N-1}}^\dagger \hat{a}_{P_N}^\dagger | \text{vac} \rangle$$

Non-interacting (delocalised) molecular orbital representation

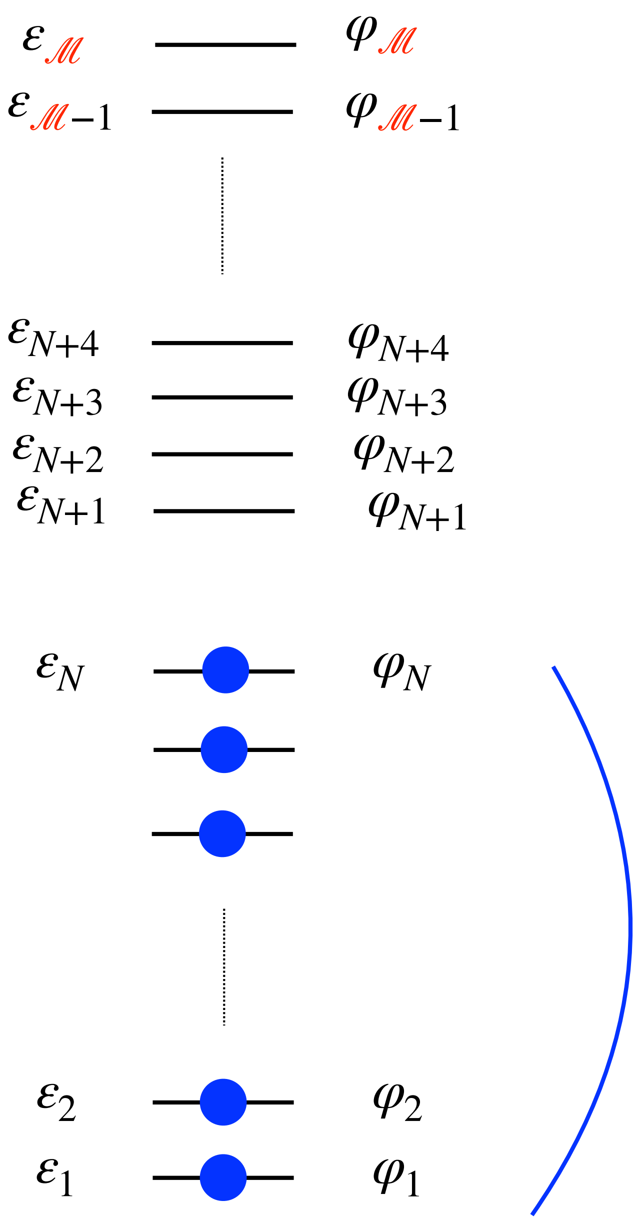


$$\hat{H} = \sum_P \varepsilon_P \hat{a}^\dagger_P \hat{a}_P$$

*Spin-orbital **occupation** operator*

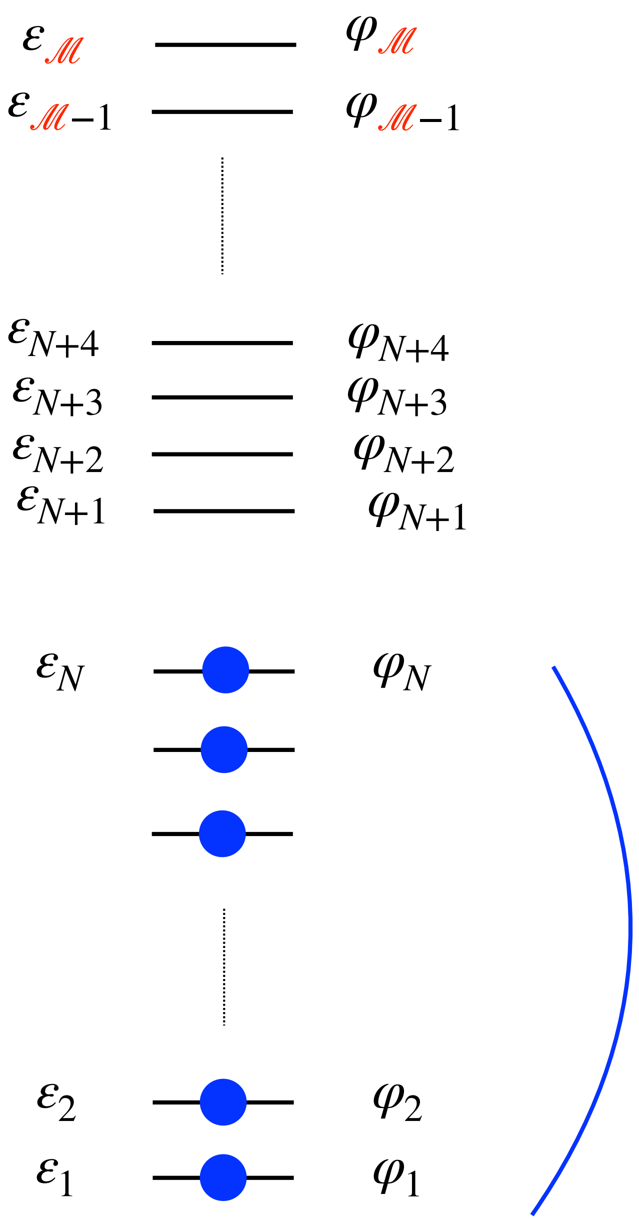
$$\hat{H}\, \hat{a}^\dagger_{P_1}\hat{a}^\dagger_{P_2}\ldots\hat{a}^\dagger_{P_{N-1}}\hat{a}^\dagger_{P_N}|\mathrm{vac}\rangle = \left(\sum_{i=1}^{N} \varepsilon_{P_i}\right)\hat{a}^\dagger_{P_1}\hat{a}^\dagger_{P_2}\ldots\hat{a}^\dagger_{P_{N-1}}\hat{a}^\dagger_{P_N}|\mathrm{vac}\rangle$$

Non-interacting (delocalised) molecular orbital representation



$$|\Psi_0\rangle = \hat{a}_1^\dagger \hat{a}_2^\dagger \ldots \hat{a}_{N-1}^\dagger \hat{a}_N^\dagger |\mathrm{vac}\rangle$$

1RDM in the molecular orbital (mo) representation



$$\gamma_{PQ}^{mo} = \langle \Psi_0 | \hat{a}_P^\dagger \hat{a}_Q | \Psi_0 \rangle$$

$$|\Psi_0\rangle = \hat{a}_1^\dagger \hat{a}_2^\dagger \ldots \hat{a}_{N-1}^\dagger \hat{a}_N^\dagger |\text{vac}\rangle$$

1RDM in the molecular orbital (mo) representation

$$\gamma_{PQ}^{mo} = \langle \Psi_0 | \hat{a}_P^\dagger \hat{a}_Q | \Psi_0 \rangle$$

where

$$|\Psi_0\rangle = \hat{a}_1^\dagger \hat{a}_2^\dagger \ldots \hat{a}_{N-1}^\dagger \hat{a}_N^\dagger |\text{vac}\rangle$$

1RDM in the molecular orbital (mo) representation

$$\gamma_{PQ}^{mo} = \langle \Psi_0 | \hat{a}_P^\dagger \hat{a}_Q | \Psi_0 \rangle \quad \text{where} \quad | \Psi_0 \rangle = \hat{a}_1^\dagger \hat{a}_2^\dagger \ldots \hat{a}_{N-1}^\dagger \hat{a}_N^\dagger | \text{vac} \rangle$$

Is φ_Q occupied in Ψ_0?

1RDM in the molecular orbital (mo) representation

$$\gamma^{mo}_{PQ} = \langle \Psi_0 | \hat{a}^\dagger_P \hat{a}_Q | \Psi_0 \rangle \quad \text{where} \quad |\Psi_0\rangle = \hat{a}^\dagger_1 \hat{a}^\dagger_2 \ldots \hat{a}^\dagger_{N-1} \hat{a}^\dagger_N |\text{vac}\rangle$$

Is φ_Q occupied in Ψ_0?

NO

$$0$$

1RDM in the molecular orbital (mo) representation

$$\gamma_{PQ}^{mo} = \langle \Psi_0 | \hat{a}_P^\dagger \hat{a}_Q | \Psi_0 \rangle \quad \text{where} \quad |\Psi_0\rangle = \hat{a}_1^\dagger \hat{a}_2^\dagger \ldots \hat{a}_{N-1}^\dagger \hat{a}_N^\dagger |\text{vac}\rangle$$

Is φ_Q occupied in Ψ_0?

NO → 0

YES → $P \overset{?}{=} Q$

1RDM in the molecular orbital (mo) representation

$$\gamma^{mo}_{PQ} = \langle \Psi_0 | \hat{a}^\dagger_P \hat{a}_Q | \Psi_0 \rangle \quad \text{where} \quad |\Psi_0\rangle = \hat{a}^\dagger_1 \hat{a}^\dagger_2 \ldots \hat{a}^\dagger_{N-1} \hat{a}^\dagger_N |\text{vac}\rangle$$

Is φ_Q occupied in Ψ_0?

NO → 0

YES → $P \overset{?}{=} Q$

NO →

If φ_P is occupied in Ψ_0 then $\hat{a}^\dagger_P \hat{a}_Q | \Psi_0 \rangle = 0$ **Pauli principle**

1RDM in the molecular orbital (mo) representation

$$\gamma_{PQ}^{mo} = \langle \Psi_0 | \hat{a}_P^\dagger \hat{a}_Q | \Psi_0 \rangle \quad \text{where} \quad |\Psi_0\rangle = \hat{a}_1^\dagger \hat{a}_2^\dagger \ldots \hat{a}_{N-1}^\dagger \hat{a}_N^\dagger |\text{vac}\rangle$$

Is φ_Q occupied in Ψ_0?

NO → 0

YES → $P \stackrel{?}{=} Q$

NO →

If φ_P is unoccupied in Ψ_0 then $\hat{a}_P^\dagger \hat{a}_Q |\Psi_0\rangle \perp |\Psi_0\rangle \Rightarrow \langle \Psi_0 | \hat{a}_P^\dagger \hat{a}_Q | \Psi_0 \rangle = 0$

1RDM in the molecular orbital (mo) representation

$$\gamma^{mo}_{PQ} = \langle \Psi_0 | \hat{a}^\dagger_P \hat{a}_Q | \Psi_0 \rangle$$ where $$|\Psi_0\rangle = \hat{a}^\dagger_1 \hat{a}^\dagger_2 \ldots \hat{a}^\dagger_{N-1} \hat{a}^\dagger_N | \text{vac} \rangle$$

Is φ_Q occupied in Ψ_0?

NO → 0

YES → $P \overset{?}{=} Q$

NO → 0

1RDM in the molecular orbital (mo) representation

$$\gamma^{mo}_{PQ} = \langle \Psi_0 | \hat{a}^\dagger_P \hat{a}_Q | \Psi_0 \rangle \quad \text{where} \quad |\Psi_0\rangle = \hat{a}^\dagger_1 \hat{a}^\dagger_2 \ldots \hat{a}^\dagger_{N-1} \hat{a}^\dagger_N |\text{vac}\rangle$$

Is φ_Q occupied in Ψ_0?

NO → 0

YES → $P \stackrel{?}{=} Q$

- **NO** → 0
- **YES** → $\hat{a}^\dagger_P \hat{a}_P |\Psi_0\rangle = |\Psi_0\rangle$

1RDM in the molecular orbital (mo) representation

$$\gamma^{mo}_{PQ} = \langle \Psi_0 | \hat{a}^\dagger_P \hat{a}_Q | \Psi_0 \rangle \quad \text{where} \quad | \Psi_0 \rangle = \hat{a}^\dagger_1 \hat{a}^\dagger_2 \ldots \hat{a}^\dagger_{N-1} \hat{a}^\dagger_N | \text{vac} \rangle$$

Is φ_Q occupied in Ψ_0?

NO → 0

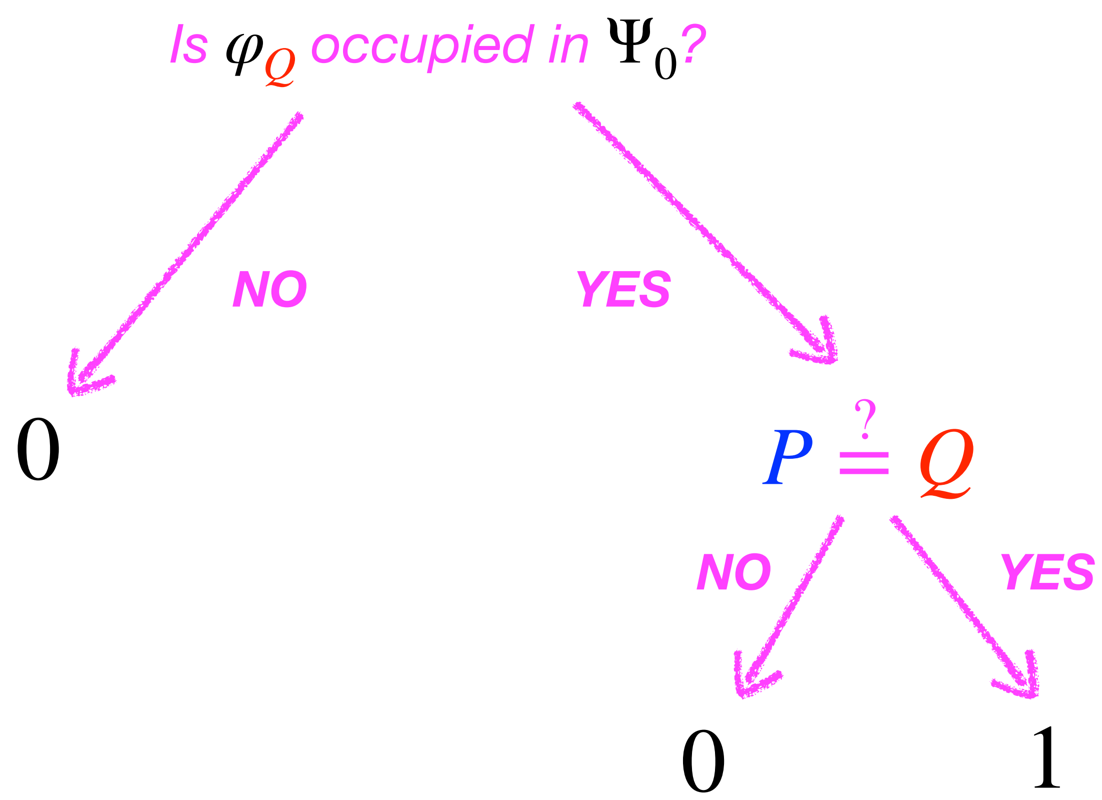
YES → $P \stackrel{?}{=} Q$

- **NO** → 0
- **YES** →

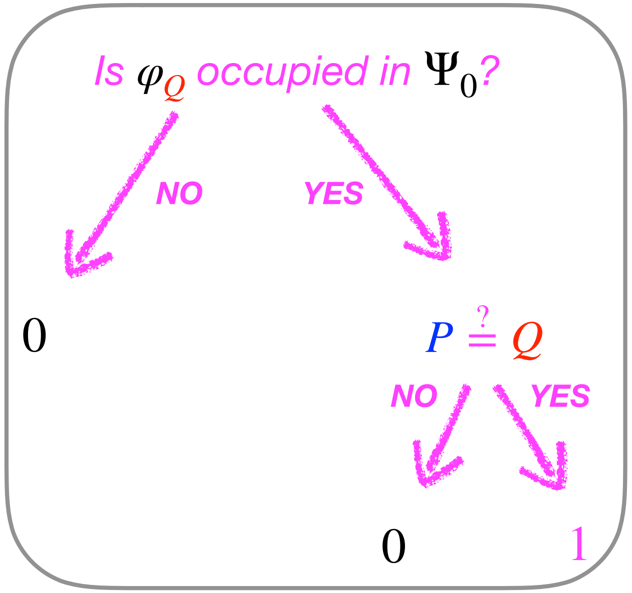
$$\gamma^{mo}_{PP} = \langle \Psi_0 | \hat{a}^\dagger_P \hat{a}_P | \Psi_0 \rangle$$
$$= \langle \Psi_0 | \Psi_0 \rangle = 1$$

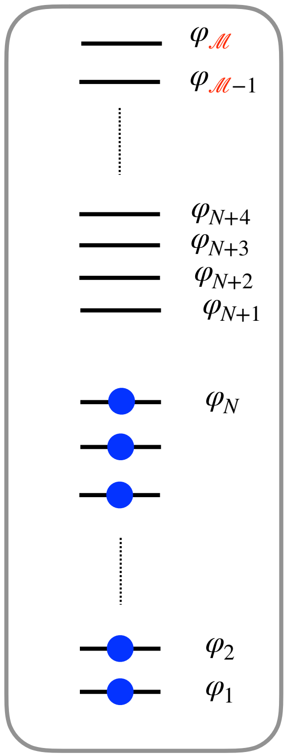
1RDM in the molecular orbital (mo) representation

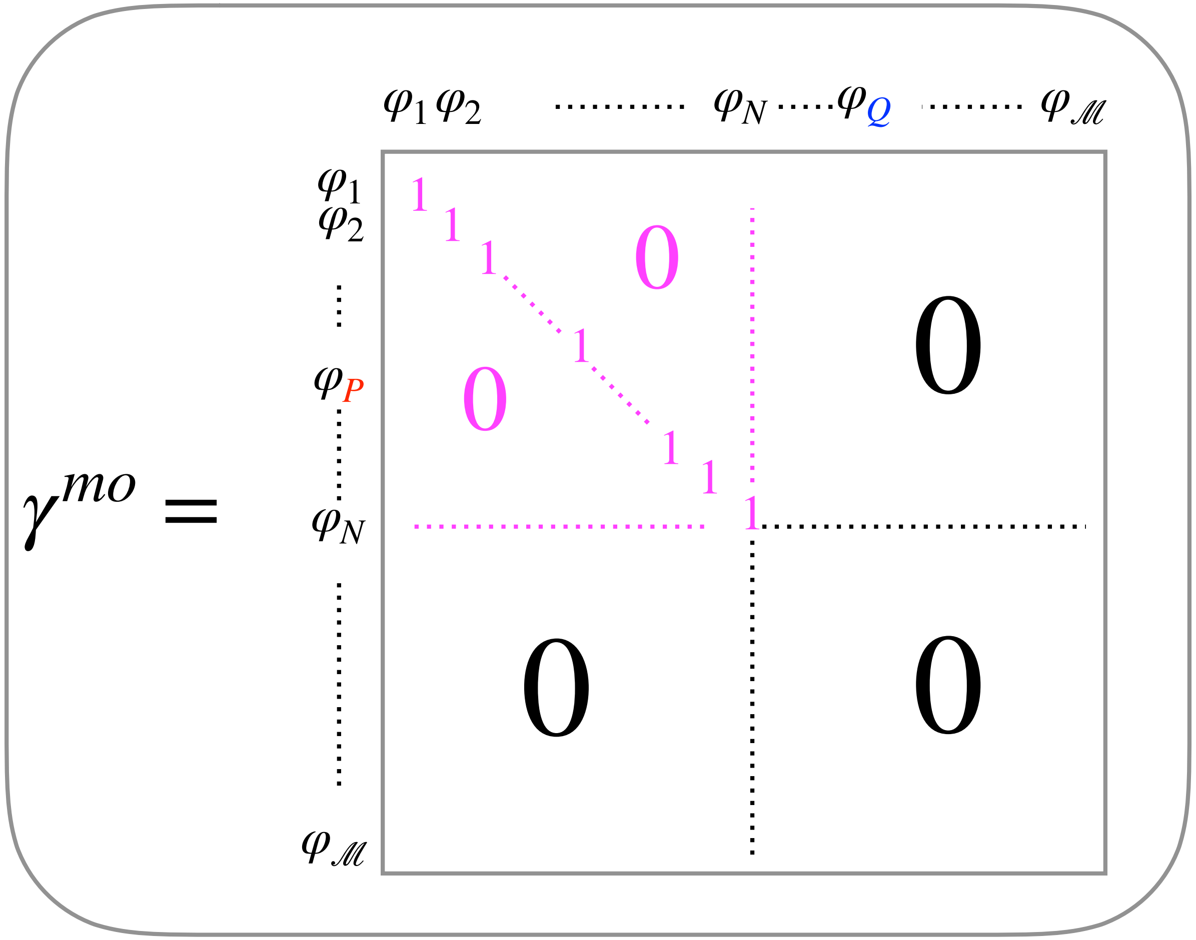
$$\gamma^{mo}_{PQ} = \langle \Psi_0 | \hat{a}^\dagger_P \hat{a}_Q | \Psi_0 \rangle \quad \text{where} \quad | \Psi_0 \rangle = \hat{a}^\dagger_1 \hat{a}^\dagger_2 \ldots \hat{a}^\dagger_{N-1} \hat{a}^\dagger_N | \text{vac} \rangle$$



1RDM in the molecular orbital (mo) representation

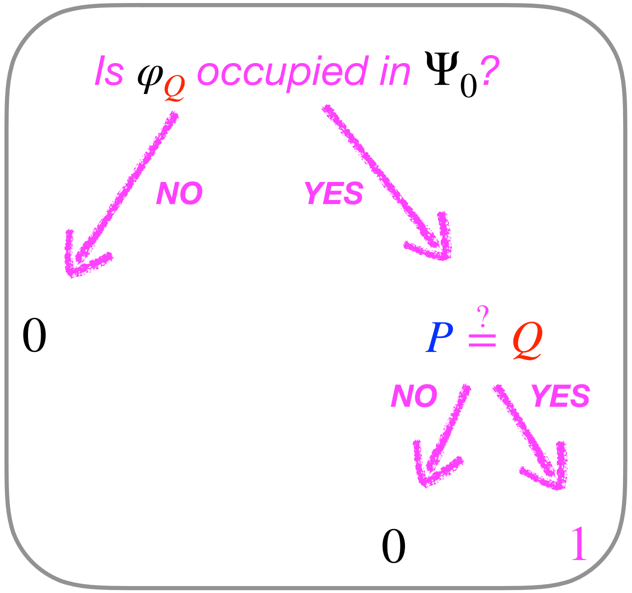


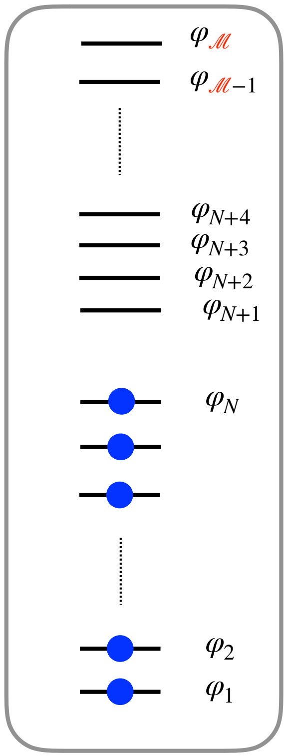


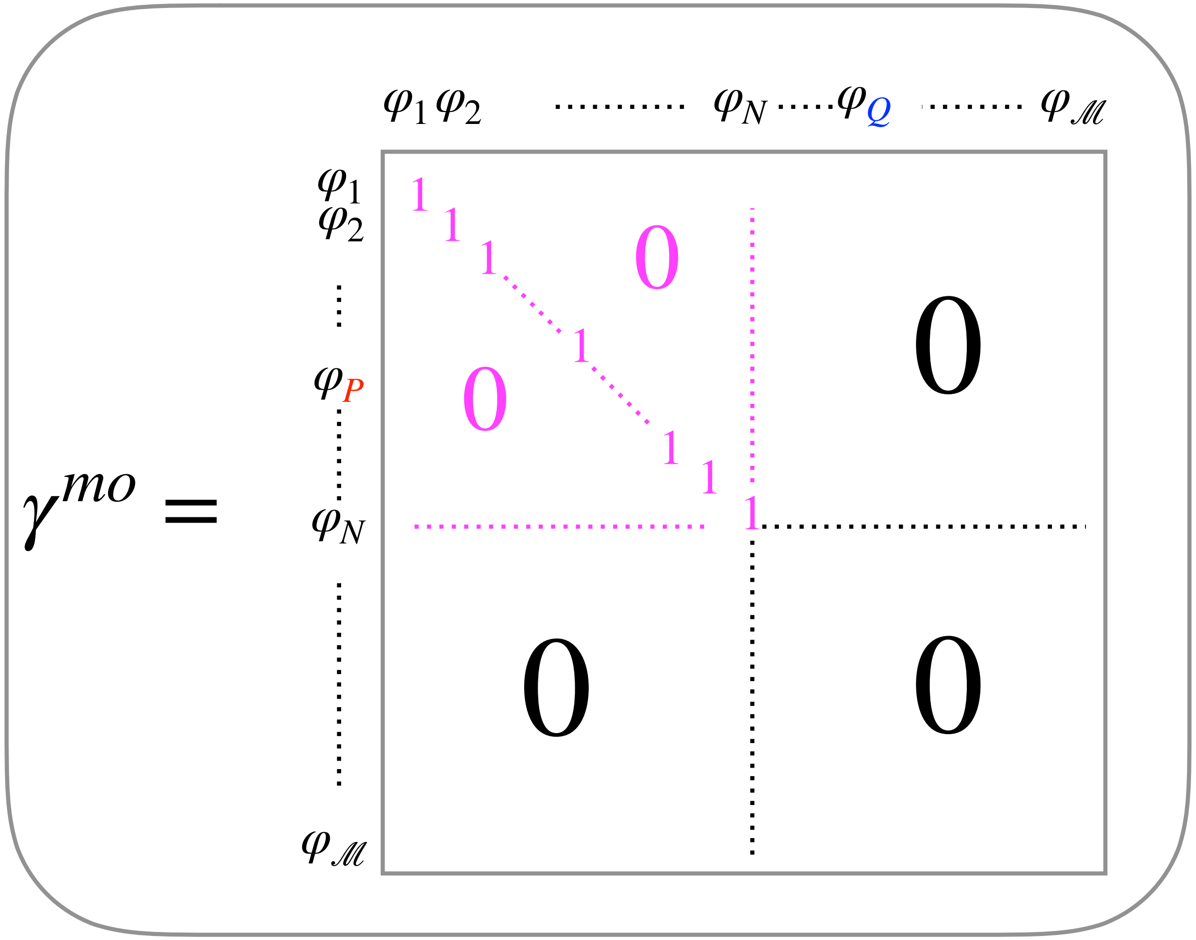


Non-interacting problem solved!

1RDM in the molecular orbital (mo) representation



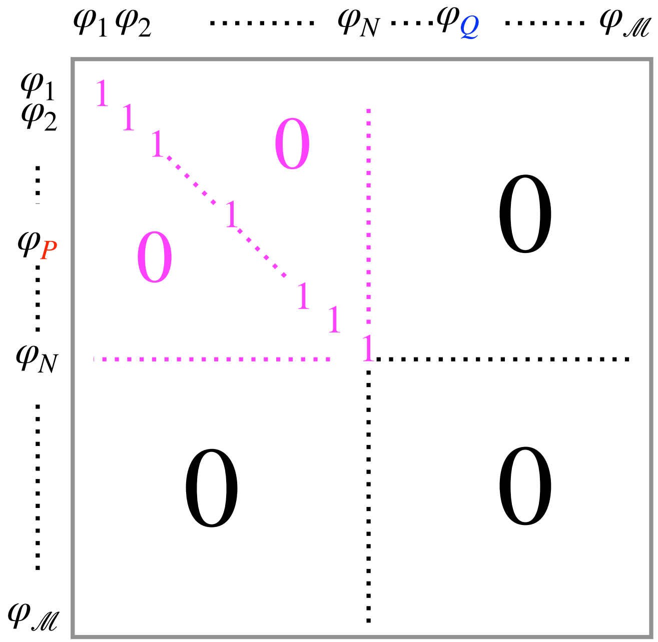




***No entanglement** between the molecular orbitals*
in the non-interacting case

Idempotency property

$\gamma^{mo} =$



$= \left[\gamma^{mo}\right]^2$

Turning to the localised picture (useless here although interesting)

$$|\varphi_P\rangle = \sum_I C_{IP} |\chi_I\rangle$$

$$|\varphi_Q\rangle = \sum_J C_{JQ} |\chi_J\rangle$$

Localised spin-orbitals

Turning to the localised picture (useless here although interesting)

$$|\varphi_P\rangle = \sum_I C_{IP} |\chi_I\rangle$$

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Localised spin-orbitals

$$\langle\varphi_P|\varphi_Q\rangle = \delta_{PQ} = \sum_{IJ} C_{IP} C_{JQ} \langle\chi_I|\chi_J\rangle$$

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Localised spin-orbitals

$$\langle\varphi_P|\varphi_Q\rangle = \delta_{PQ} = \sum_{IJ} C_{IP} C_{JQ} \langle\chi_I|\chi_J\rangle$$

Orthonormalisation *procedure*

$$\delta_{IJ}$$

Turning to the localised picture (useless here although interesting)

$$|\varphi_P\rangle = \sum_I C_{IP} |\chi_I\rangle$$

$$|\varphi_Q\rangle = \sum_J C_{JQ} |\chi_J\rangle$$

Localised spin-orbitals

Molecular orbital coefficients **matrix**

$$\langle\varphi_P|\varphi_Q\rangle = \delta_{PQ} = \sum_{IJ} C_{IP}C_{JQ}\,\delta_{IJ}$$

$$= \sum_I C_{IP}C_{IQ} = \sum_I \left[\mathbf{C}^{\mathrm{T}}\right]_{PI} \left[\mathbf{C}\right]_{IQ}$$

$$= \left[\mathbf{C}^{\mathrm{T}}\mathbf{C}\right]_{PQ}$$

Turning to the localised picture (useless here although interesting)

$$|\varphi_P\rangle = \sum_I C_{IP} |\chi_I\rangle$$

$$|\varphi_Q\rangle = \sum_J C_{JQ} |\chi_J\rangle$$

Localised spin-orbitals

Unitary transformation

from the delocalised to localised pictures

$$\mathbf{C}^{-1} = \mathbf{C}^{\mathrm{T}}$$

$$\delta_{PQ} = \left[\mathbf{C}^{\mathrm{T}}\mathbf{C}\right]_{PQ}$$

Turning to the localised picture (useless here although interesting)

$$\gamma_{PQ}^{mo} = \langle \Psi_0 | \hat{a}_P^\dagger \hat{a}_Q | \Psi_0 \rangle$$

$$\hat{a}_P^\dagger = \sum_I C_{IP} \hat{c}_I^\dagger$$

$$\hat{a}_Q = \sum_J C_{JQ} \hat{c}_J$$

$$\gamma_{PQ}^{mo} = \sum_{IJ} C_{IP} C_{JQ} \langle \Psi_0 | \hat{c}_I^\dagger \hat{c}_J | \Psi_0 \rangle$$

Turning to the localised picture (useless here although interesting)

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1RDM in the **localised** representation

$$\gamma_{PQ}^{mo} = \sum_{IJ} C_{IP} C_{JQ} \langle \Psi_0 | \hat{c}_I^\dagger \hat{c}_J | \Psi_0 \rangle$$

$$\gamma_{IJ}^{loc}$$

Turning to the localised picture (useless here although interesting)

$$\gamma_{PQ}^{mo} = \langle \Psi_0 | \hat{a}_P^\dagger \hat{a}_Q | \Psi_0 \rangle$$

$$\hat{a}_P^\dagger = \sum_I C_{IP} \hat{c}_I^\dagger$$

$$\hat{a}_Q = \sum_J C_{JQ} \hat{c}_J$$

$$\gamma_{PQ}^{mo} = \sum_{IJ} C_{IP} C_{JQ} \gamma_{IJ}^{loc} = \sum_{IJ} [\mathbf{C}^{\mathrm{T}}]_{PI} \gamma_{IJ}^{loc} [\mathbf{C}]_{JQ}$$

Turning to the localised picture (useless here although interesting)

$$\gamma_{PQ}^{mo} = \langle \Psi_0 | \hat{a}_P^\dagger \hat{a}_Q | \Psi_0 \rangle$$

$$\hat{a}_P^\dagger = \sum_I C_{IP}\, \hat{c}_I^\dagger$$

$$\hat{a}_Q = \sum_J C_{JQ}\, \hat{c}_J$$

$$\gamma_{PQ}^{mo} = \sum_{IJ} C_{IP} C_{JQ}\, \gamma_{IJ}^{loc} = \sum_{IJ} \left[\mathbf{C}^{\mathrm{T}}\right]_{PI} \gamma_{IJ}^{loc} \left[\mathbf{C}\right]_{JQ}$$

$$= \left[\mathbf{C}^{\mathrm{T}} \gamma^{loc} \mathbf{C}\right]_{PQ}$$

Turning to the localised picture (useless here although interesting)

$$\boxed{\gamma^{loc} = \mathbf{C}\gamma^{mo}\mathbf{C}^{\mathrm{T}}}$$

$$\mathbf{C} \times \longrightarrow \quad \uparrow \quad \longleftarrow \times \mathbf{C}^{\mathrm{T}}$$

$$\mathbf{C}^{-1} = \mathbf{C}^{\mathrm{T}}$$

$$\gamma^{mo} = \mathbf{C}^{\mathrm{T}}\gamma^{loc}\mathbf{C}$$

$$\uparrow$$

$$\gamma^{mo}_{PQ} = \left[\mathbf{C}^{\mathrm{T}}\gamma^{loc}\mathbf{C}\right]_{PQ}$$

Turning to the localised picture (useless here although interesting)

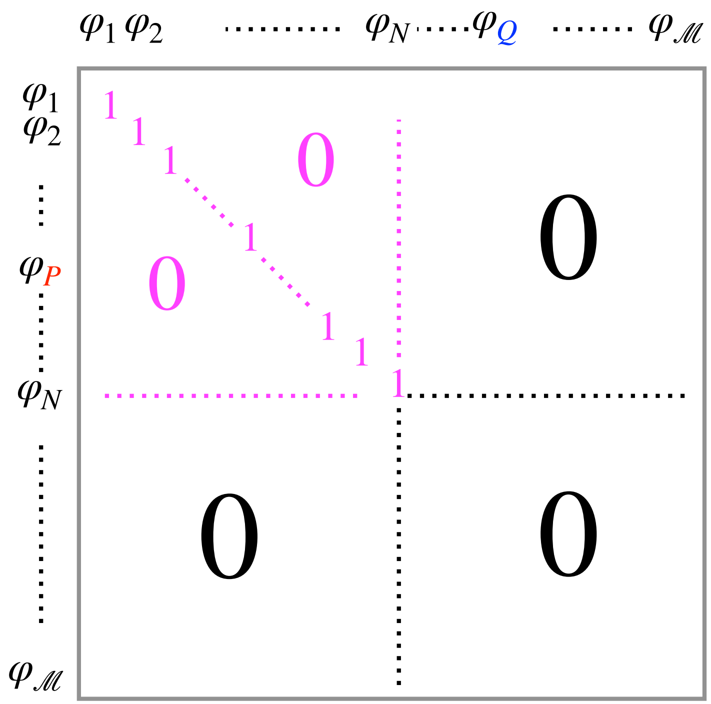
$$\gamma^{loc} = \mathbf{C}\gamma^{mo}\mathbf{C}^{\mathrm{T}}$$

↓

$$\gamma_{IJ}^{loc} = \sum_{PQ} C_{IP}\,\gamma_{PQ}^{mo}\,C_{JQ}$$

Turning to the localised picture (useless here although interesting)

$$\gamma^{mo} =$$



$$\gamma_{IJ}^{loc} = \sum_{PQ} C_{IP}\,\gamma_{PQ}^{mo}\,C_{JQ} = \sum_{P}^{\textbf{occupied}\ spin-MOs} C_{IP}C_{JP}$$

Turning to the localised picture (useless here although interesting)

Note that $\sum_{P}^{\text{all spin-MOs}} \underbrace{\langle \chi_I | \varphi_P \rangle}_{C_{IP}} \underbrace{\langle \varphi_P | \chi_J \rangle}_{C_{JP}} = \langle \chi_I | \chi_J \rangle = \delta_{IJ}$

$$\gamma_{IJ}^{loc} = \sum_{PQ} C_{IP}\, \gamma_{PQ}^{mo}\, C_{JQ} = \sum_{P}^{\text{occupied spin-MOs}} C_{IP} C_{JP}$$

Turning to the localised picture (useless here although interesting)

occupied+unoccupied

Note that $\sum_{P}^{\text{all } spin-MOs} \langle \chi_I | \varphi_P \rangle \langle \varphi_P | \chi_J \rangle = \langle \chi_I | \chi_J \rangle = \delta_{IJ}$

where $\langle \chi_I | \varphi_P \rangle = C_{IP}$ and $\langle \varphi_P | \chi_J \rangle = C_{JP}$

$$\gamma_{IJ}^{loc} = \sum_{PQ} C_{IP}\, \gamma_{PQ}^{mo}\, C_{JQ} = \sum_{P}^{occupied\ spin-MOs} C_{IP} C_{JP}$$

Turning to the localised picture (useless here although interesting)

Resolution of the identity

Note that $\sum_{P}^{\text{all spin-MOs}} \underbrace{\langle \chi_I | \varphi_P \rangle}_{C_{IP}} \underbrace{\langle \varphi_P | \chi_J \rangle}_{C_{JP}} = \langle \chi_I | \chi_J \rangle = \delta_{IJ}$

$$\gamma_{IJ}^{loc} = \sum_{PQ} C_{IP}\, \gamma_{PQ}^{mo}\, C_{JQ} = \sum_{P}^{\text{occupied spin-MOs}} C_{IP} C_{JP}$$

Turning to the localised picture (useless here although interesting)

$$\gamma^{loc} = \mathbf{C}\gamma^{mo}\mathbf{C}^{\mathrm{T}}$$ → ***Not diagonal!***

$$\gamma_{IJ}^{loc} = \langle \hat{c}_I^\dagger \hat{c}_J \rangle_{\Psi_0} = \sum_{P}^{\textbf{occupied}\ spin-MOs} C_{IP} C_{JP} \neq \delta_{IJ}$$

Turning to the localised picture (useless here although interesting)

$$\gamma^{loc} = \mathbf{C}\gamma^{mo}\mathbf{C}^{\mathrm{T}}$$

← ***Not diagonal!***

$$\gamma_{IJ}^{loc} = \langle \hat{c}_I^\dagger \hat{c}_J \rangle_{\Psi_0} = \sum_{P}^{occupied\ spin-MOs} C_{IP} C_{JP} \neq \delta_{IJ}$$

*Any localised spin-orbital χ_I is **entangled** with the other spin-orbitals χ_J*

Turning to the localised picture (useless here although interesting)

$$\boxed{\gamma^{loc} = \mathbf{C}\gamma^{mo}\mathbf{C}^{\mathrm{T}}}$$ ⟵ ***Not diagonal!***

$$\gamma^{loc}_{IJ} = \langle \hat{c}^{\dagger}_{I}\hat{c}_{J}\rangle_{\Psi_0} = \sum_{P}^{occupied\ spin-MOs} C_{IP}C_{JP} \neq \delta_{IJ}$$

Any localised spin-orbital χ_I *is* ***entangled*** *with the other spin-orbitals* χ_J

unlike in the delocalised molecular orbital space!

Turning to the localised picture (useless here although interesting)

$$\gamma^{loc} = \mathbf{C}\gamma^{mo}\mathbf{C}^{\mathrm{T}}$$

$$\left[\gamma^{loc}\right]^2 = \mathbf{C}\gamma^{mo}\mathbf{C}^{\mathrm{T}}\mathbf{C}\gamma^{mo}\mathbf{C}^{\mathrm{T}}$$

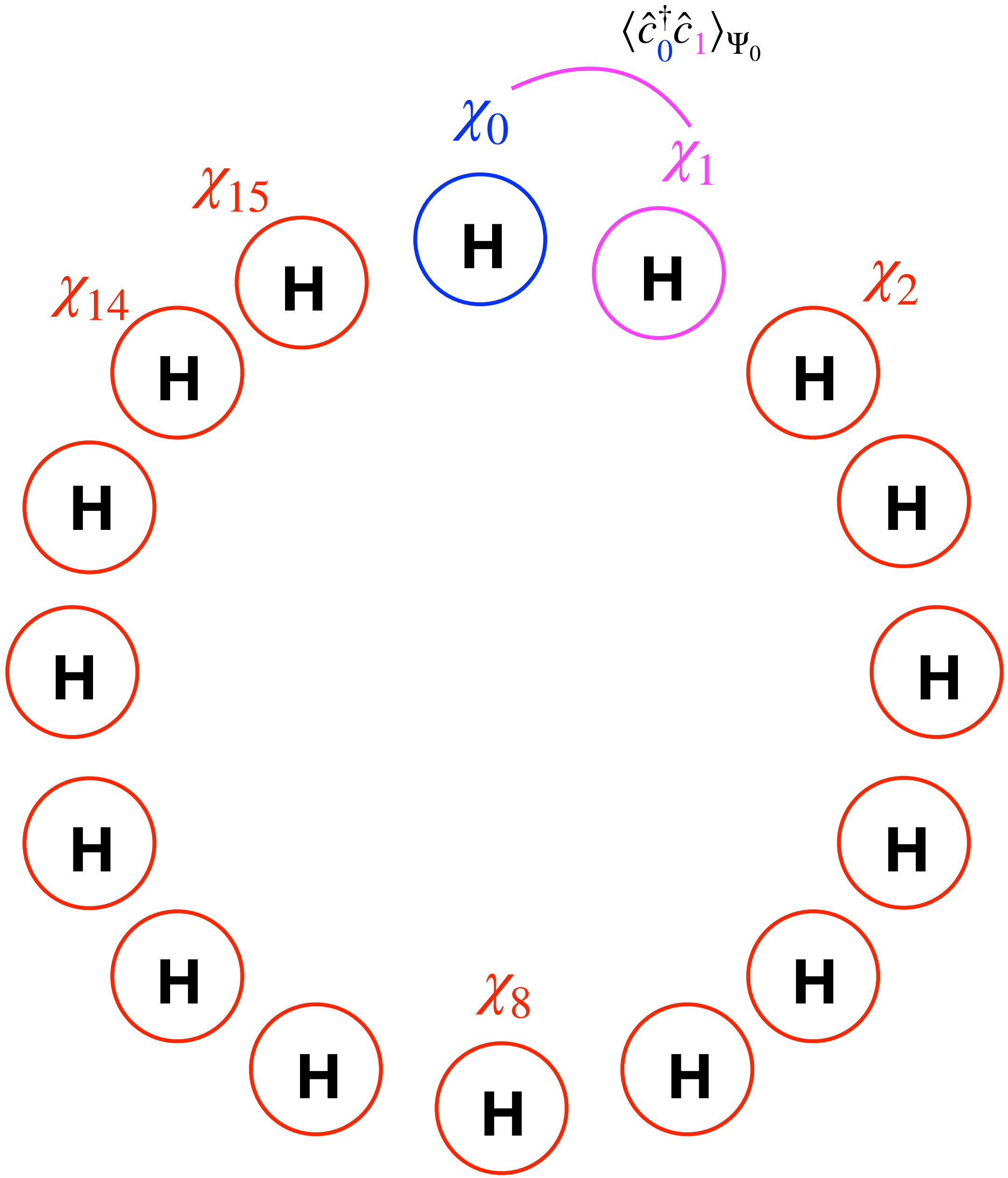
$$\mathbf{C}^{-1} = \mathbf{C}^{\mathrm{T}}$$

$$= \mathbf{C}\left[\gamma^{mo}\right]^2\mathbf{C}^{\mathrm{T}}$$

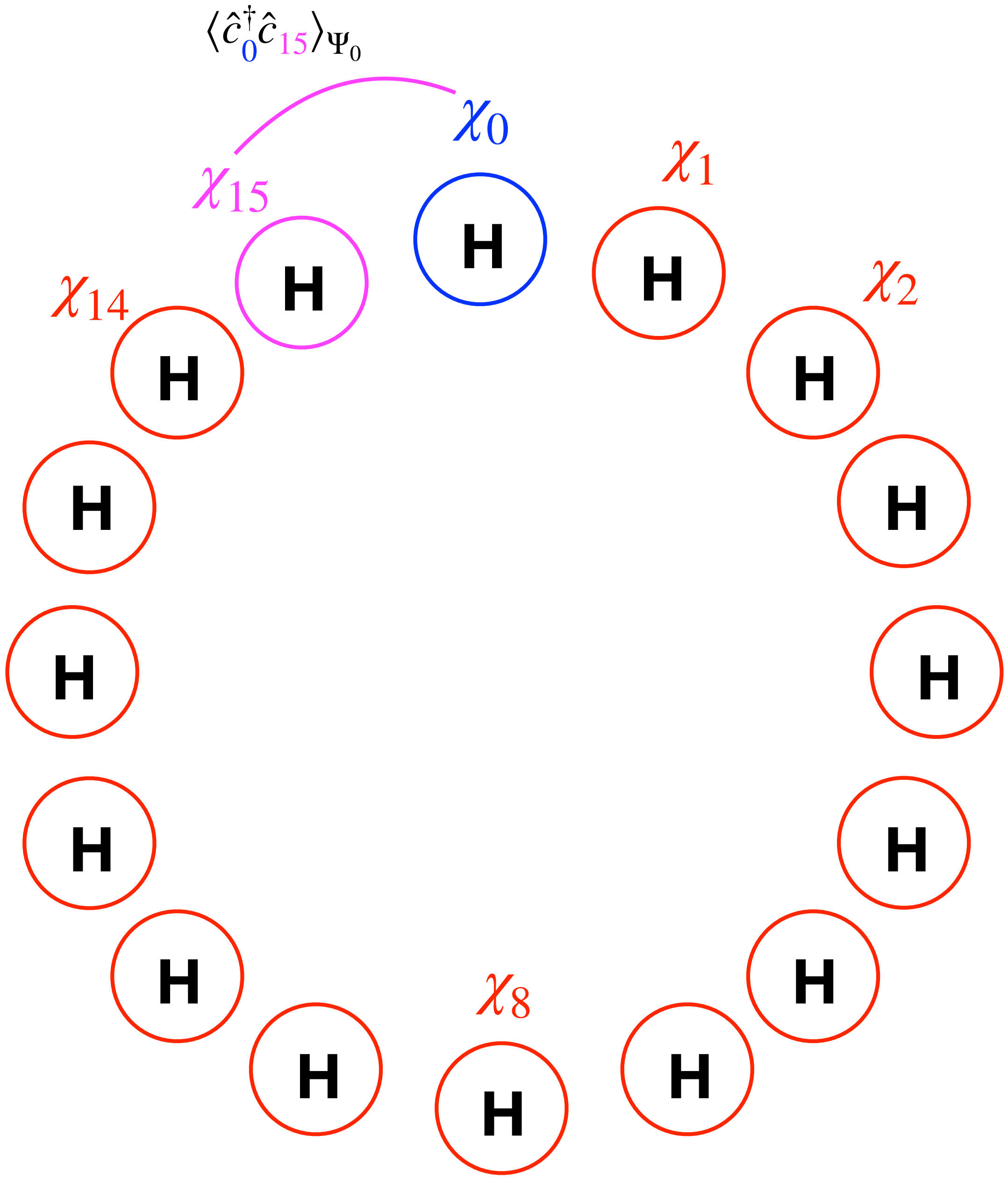
$$= \mathbf{C}\gamma^{mo}\mathbf{C}^{\mathrm{T}}$$

$$= \gamma^{loc}$$ ***Idempotent***

Prototypical ring of $L = 16$ hydrogen atoms

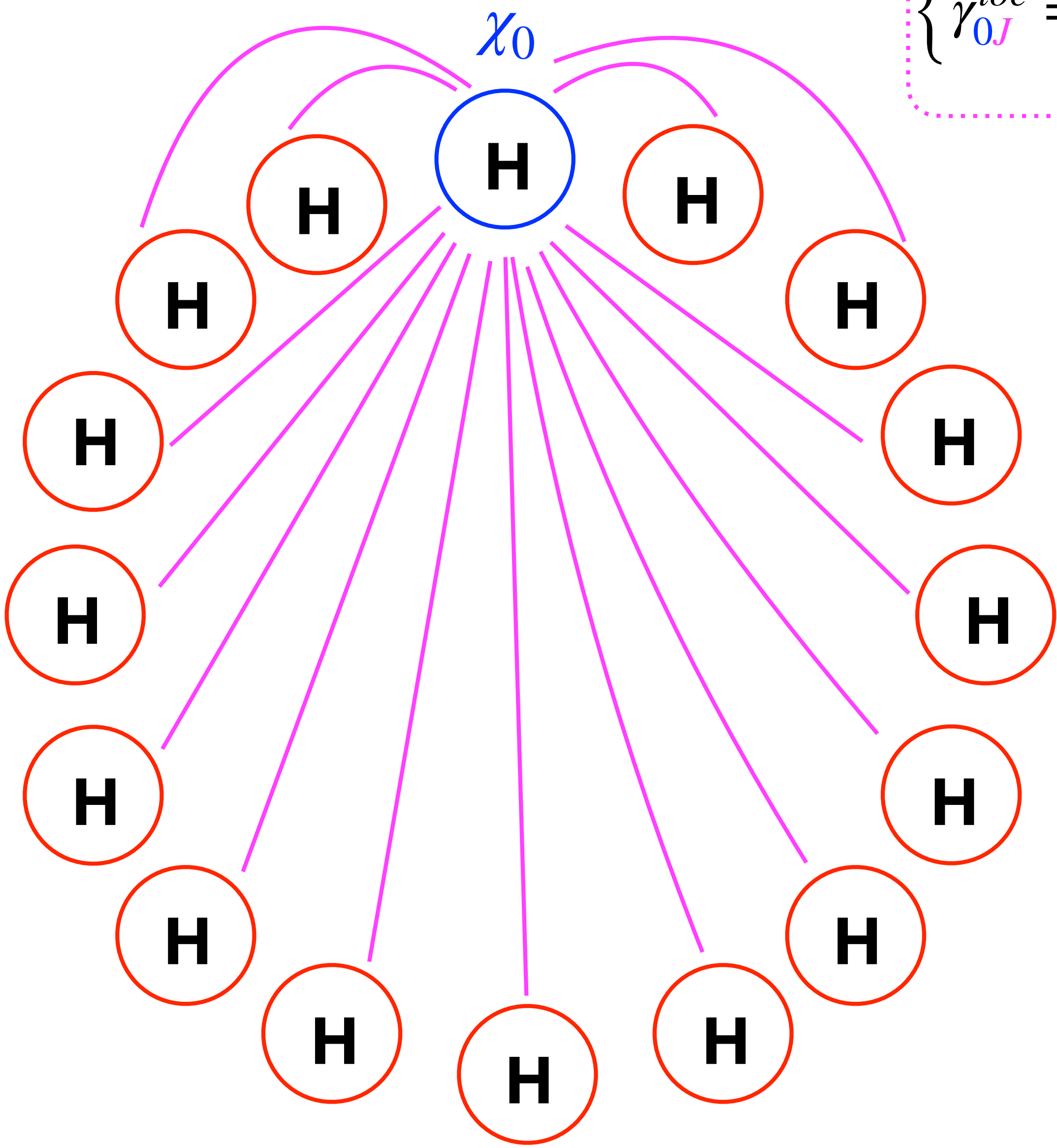


Prototypical ring of $L = 16$ hydrogen atoms

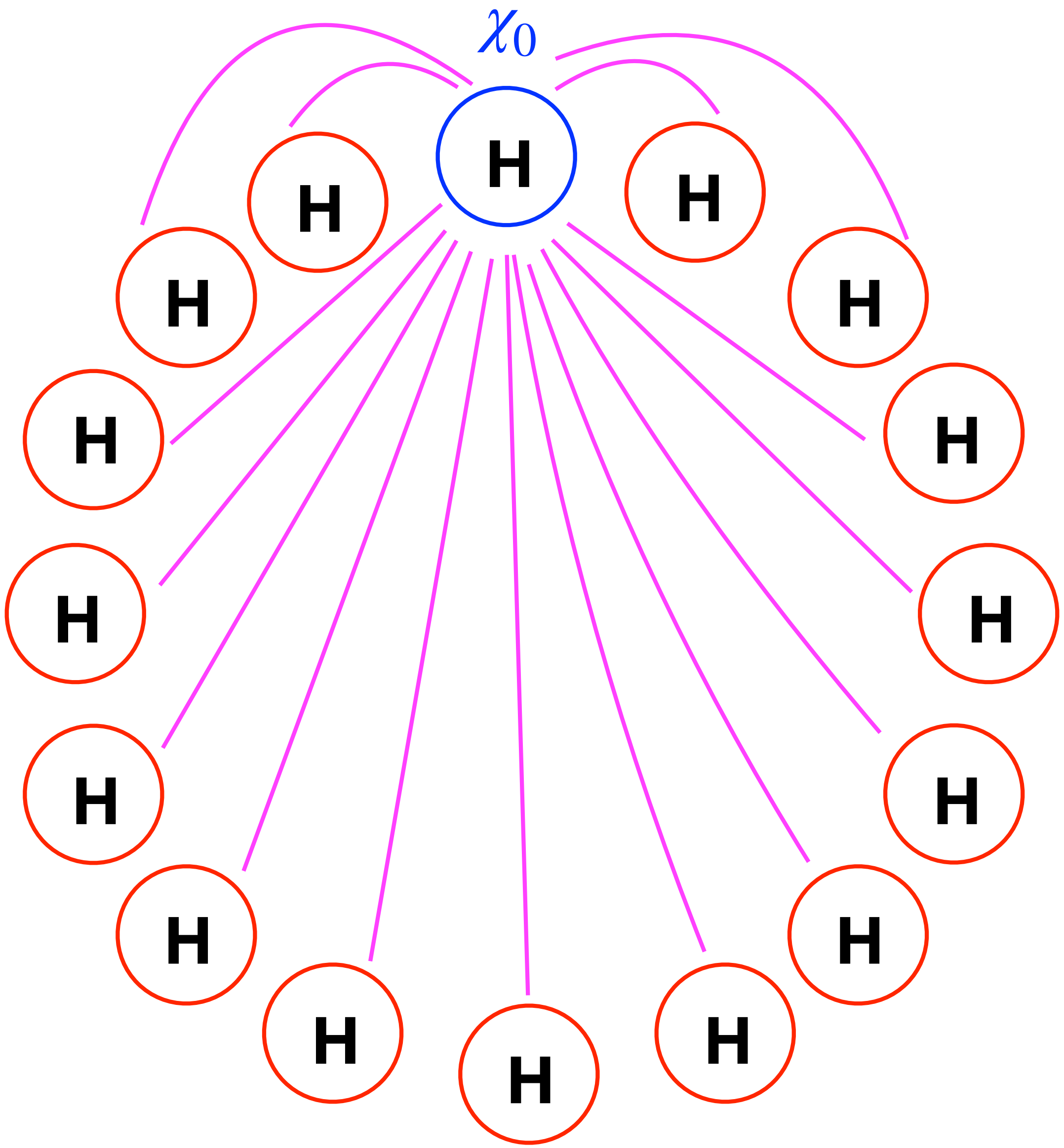


Prototypical ring of $L = 16$ hydrogen atoms

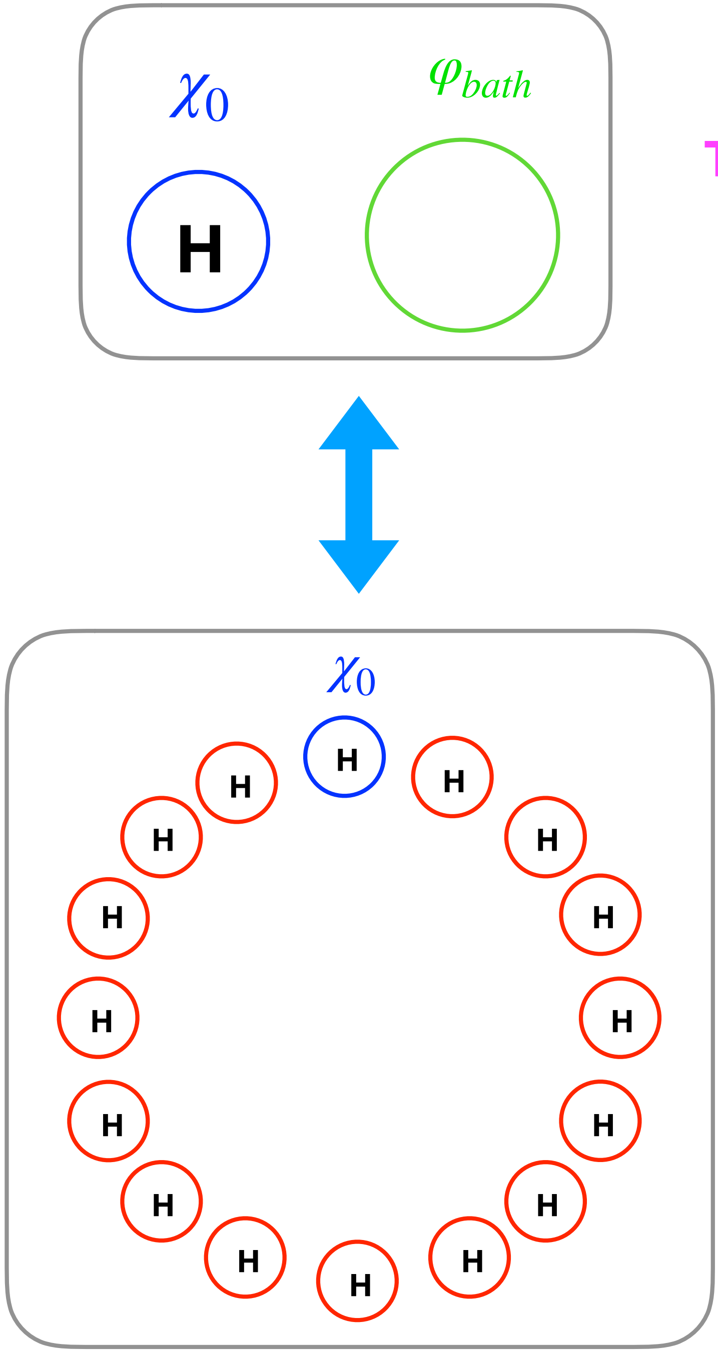
$$\left\{ \gamma_{0J}^{loc} = \langle \hat{c}_0^\dagger \hat{c}_J \rangle_{\Psi_0} \right\}_{1 \leq J \leq 15}$$



To-be-embedded (so-called ***impurity***)
localised orbital



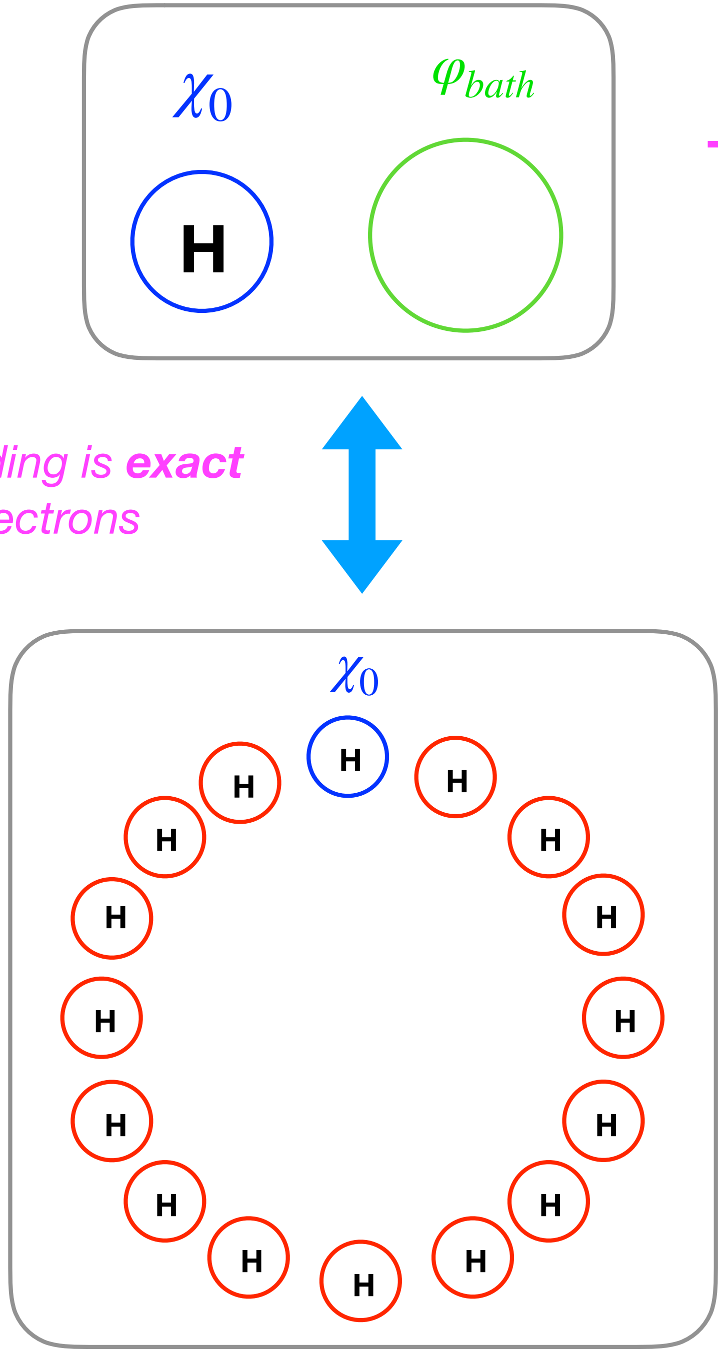
Exact density matrix functional embedding



Two-electron system

N-electron system

Exact density matrix functional embedding



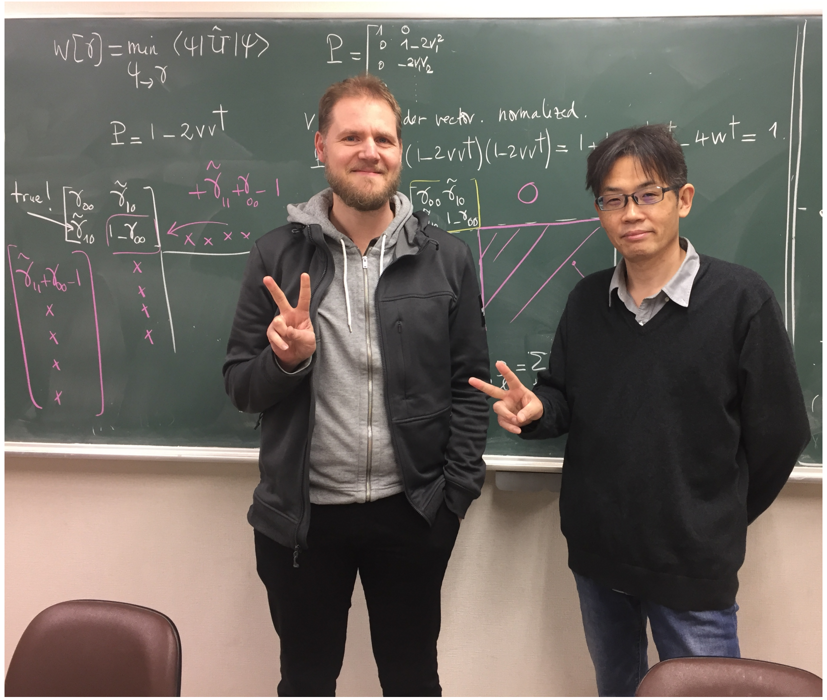
Two-electron system

*Let's prove that the embedding is **exact** for **non-interacting** electrons*

N**-electron system**

The “Householder embedding” project

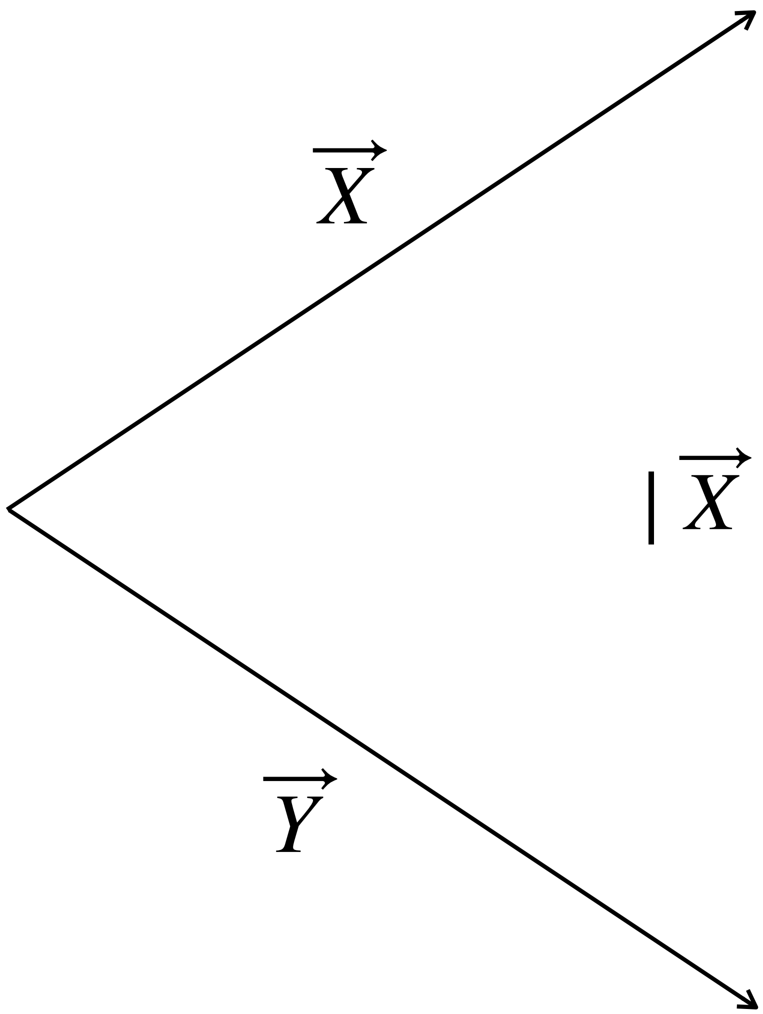
From left to right: ***S. Sekaran*** *(Strasbourg, France),*
M. Saubanère *(Montpellier, France),*
L. Mazouin *(Strasbourg, France), and E.F.*



E.F. and ***M. Tsuchiizu*** *(Nara, Japan).*

A. S. Householder, J. ACM 5, 339 (1958).
S. Sekaran, M. Tsuchiizu, M. Saubanère, and E. Fromager, Phys. Rev. B ***104****, 035121 (2021).*

The Householder transformation



$$|\overrightarrow{X}| = |\overrightarrow{Y}|$$

A. S. Householder, J. ACM 5, 339 (1958).
S. Sekaran, M. Tsuchiizu, M. Saubanère, and E. Fromager, Phys. Rev. B ***104****, 035121 (2021).*

The Householder transformation

$$\overrightarrow{X} = \sum_{i \geq 0} X_i \, \overrightarrow{e_i} \equiv \begin{bmatrix} X_0 \\ X_1 \\ \vdots \\ X_i \\ \vdots \end{bmatrix} \overset{notation}{=} \mathbf{X}$$

A. S. Householder, J. ACM 5, 339 (1958).
S. Sekaran, M. Tsuchiizu, M. Saubanère, and E. Fromager, Phys. Rev. B **104**, 035121 (2021).

The Householder transformation

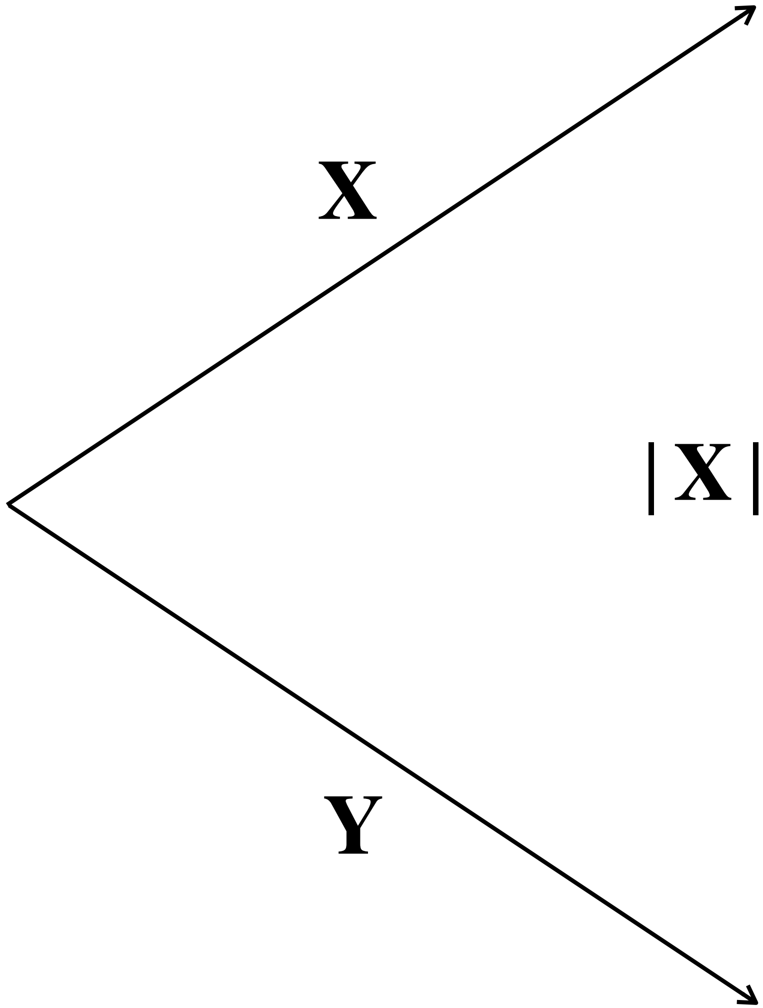
$$\vec{X} \, . \, \vec{Y} = \sum_{i \geq 0} X_i Y_i = \begin{bmatrix} X_0 & X_1 & \dots & X_i & \dots \end{bmatrix} \times \begin{bmatrix} Y_0 \\ Y_1 \\ \vdots \\ Y_i \\ \vdots \end{bmatrix}$$

$$= \mathbf{X}^{\mathrm{T}} \mathbf{Y}$$

A. S. Householder, J. ACM 5, 339 (1958).

*S. Sekaran, M. Tsuchiizu, M. Saubanère, and E. Fromager, Phys. Rev. B **104**, 035121 (2021).*

The Householder transformation

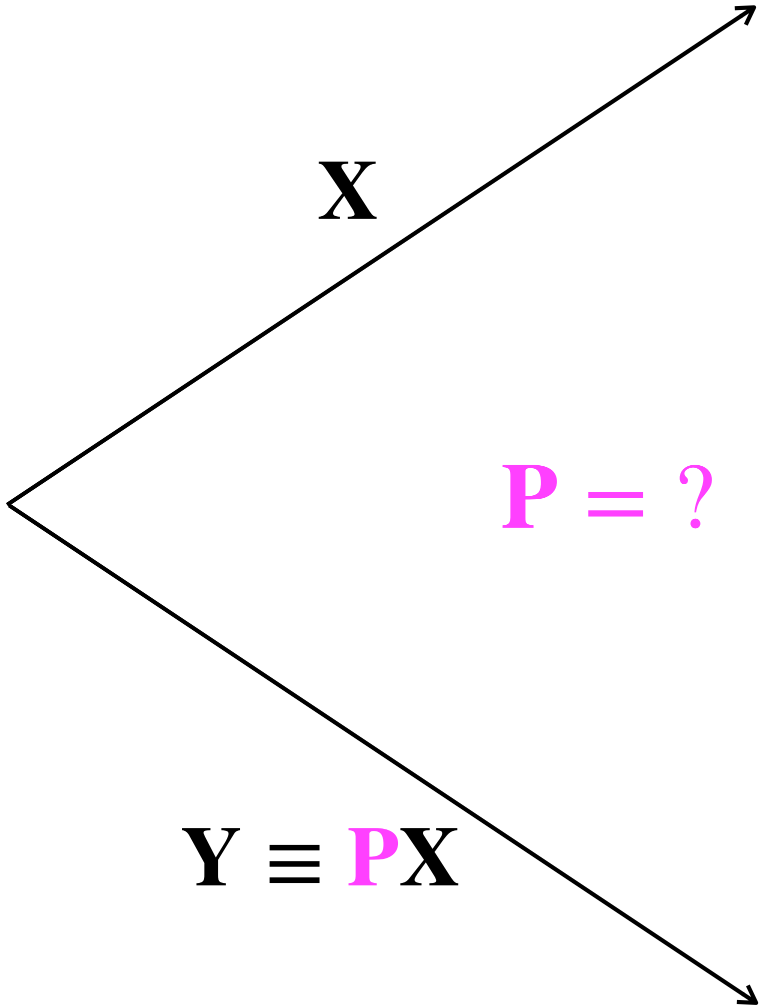


$$|\mathbf{X}| = |\mathbf{Y}|$$

A. S. Householder, J. ACM 5, 339 (1958).

*S. Sekaran, M. Tsuchiizu, M. Saubanère, and E. Fromager, Phys. Rev. B **104**, 035121 (2021).*

The Householder transformation

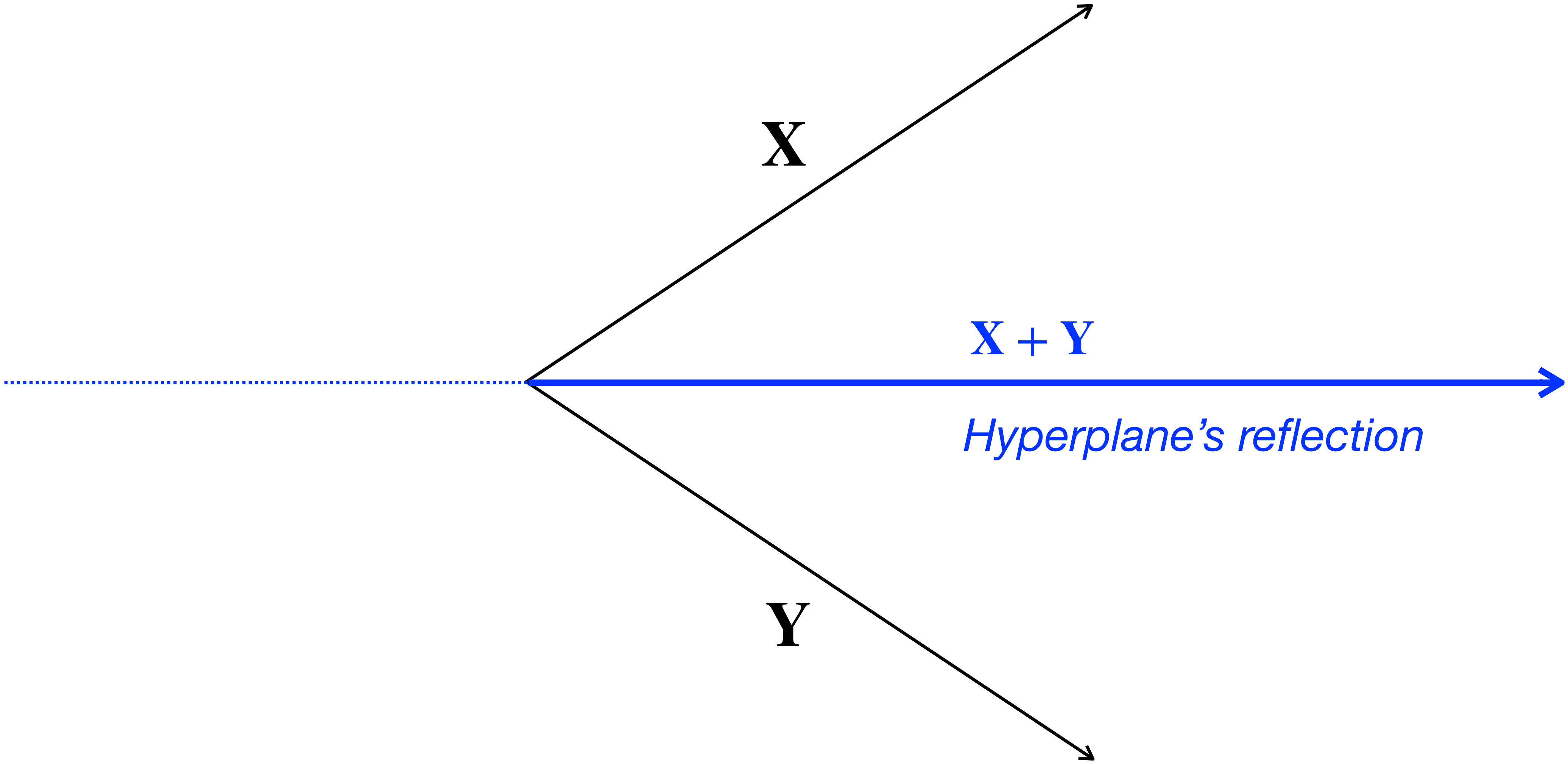


$\mathbf{P} = ?$ *Unitary square matrix*

$\mathbf{Y} \equiv \mathbf{P}\mathbf{X}$

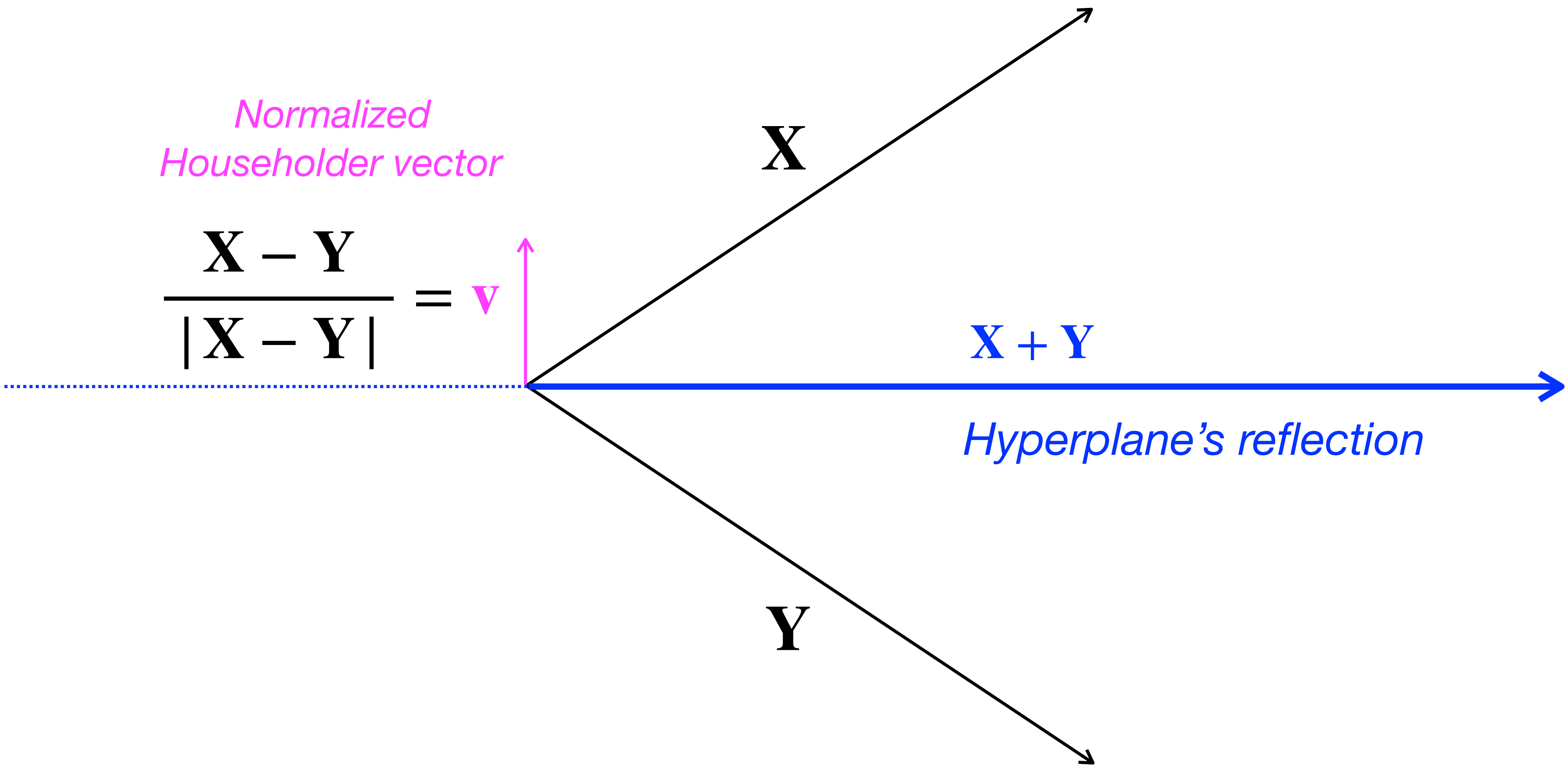
A. S. Householder, J. ACM 5, 339 (1958).
*S. Sekaran, M. Tsuchiizu, M. Saubanère, and E. Fromager, Phys. Rev. B **104**, 035121 (2021).*

The Householder transformation



A. S. Householder, J. ACM 5, 339 (1958).
*S. Sekaran, M. Tsuchiizu, M. Saubanère, and E. Fromager, Phys. Rev. B **104**, 035121 (2021).*

The Householder transformation



A. S. Householder, J. ACM 5, 339 (1958).
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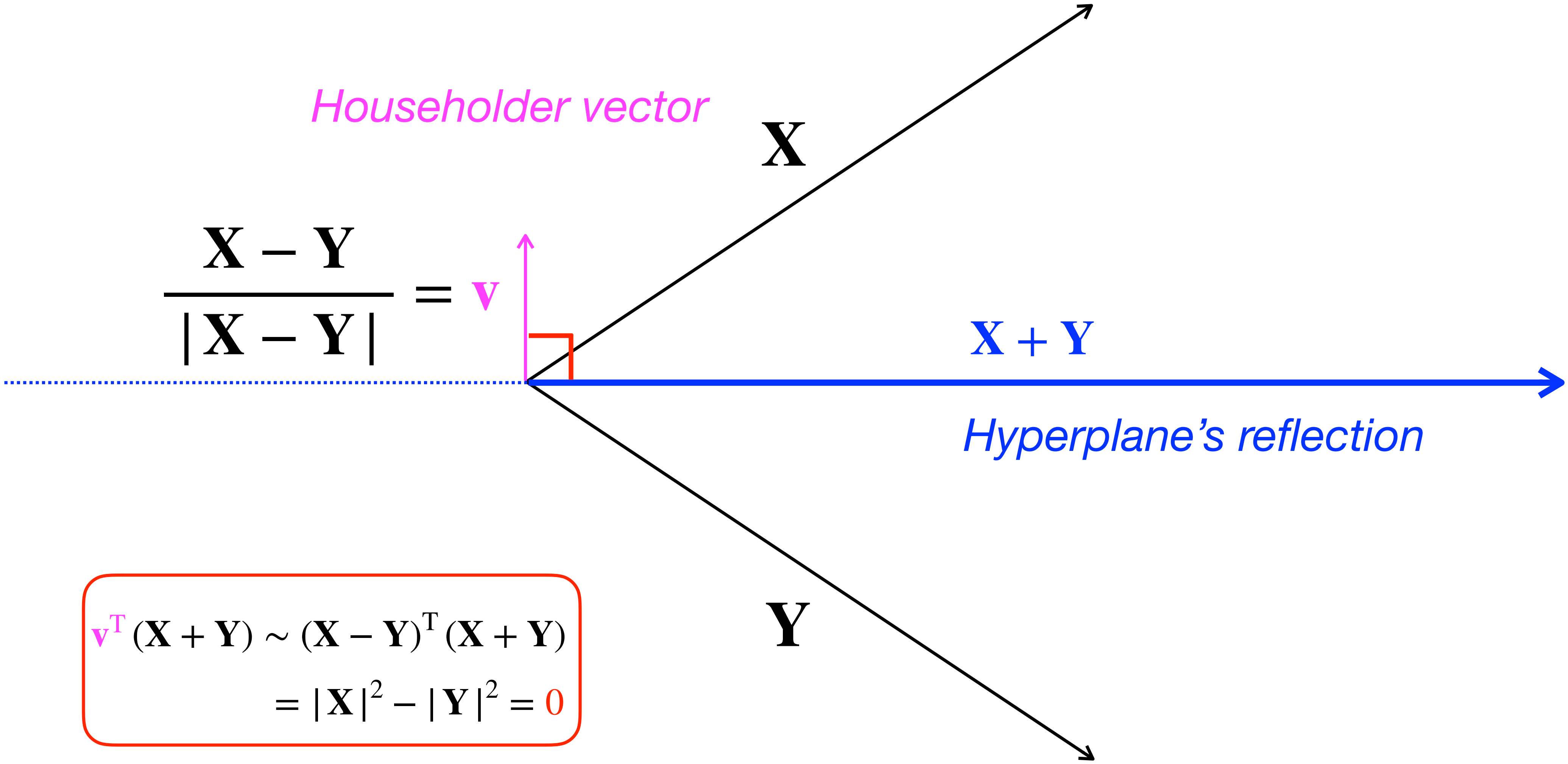
The Householder transformation

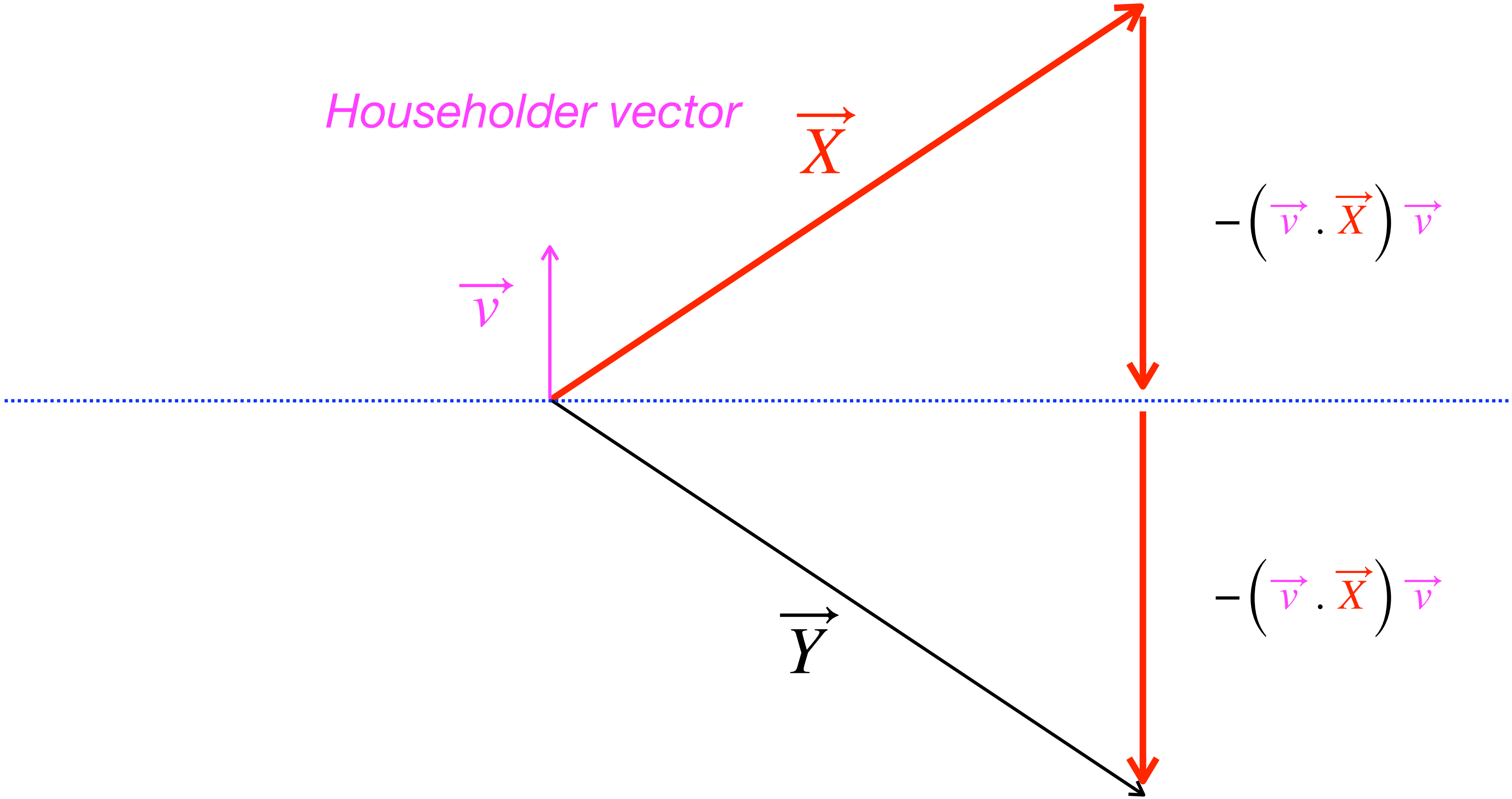


$$\frac{\mathbf{X}-\mathbf{Y}}{|\mathbf{X}-\mathbf{Y}|} = \mathbf{v}$$

$$\mathbf{v}^{\mathrm{T}}(\mathbf{X}+\mathbf{Y}) \sim (\mathbf{X}-\mathbf{Y})^{\mathrm{T}}(\mathbf{X}+\mathbf{Y})$$
$$= |\mathbf{X}|^2 - |\mathbf{Y}|^2 = 0$$

A. S. Householder, J. ACM 5, 339 (1958).
S. Sekaran, M. Tsuchiizu, M. Saubanère, and E. Fromager, Phys. Rev. B ***104****, 035121 (2021).*

The Householder transformation



A. S. Householder, J. ACM 5, 339 (1958).
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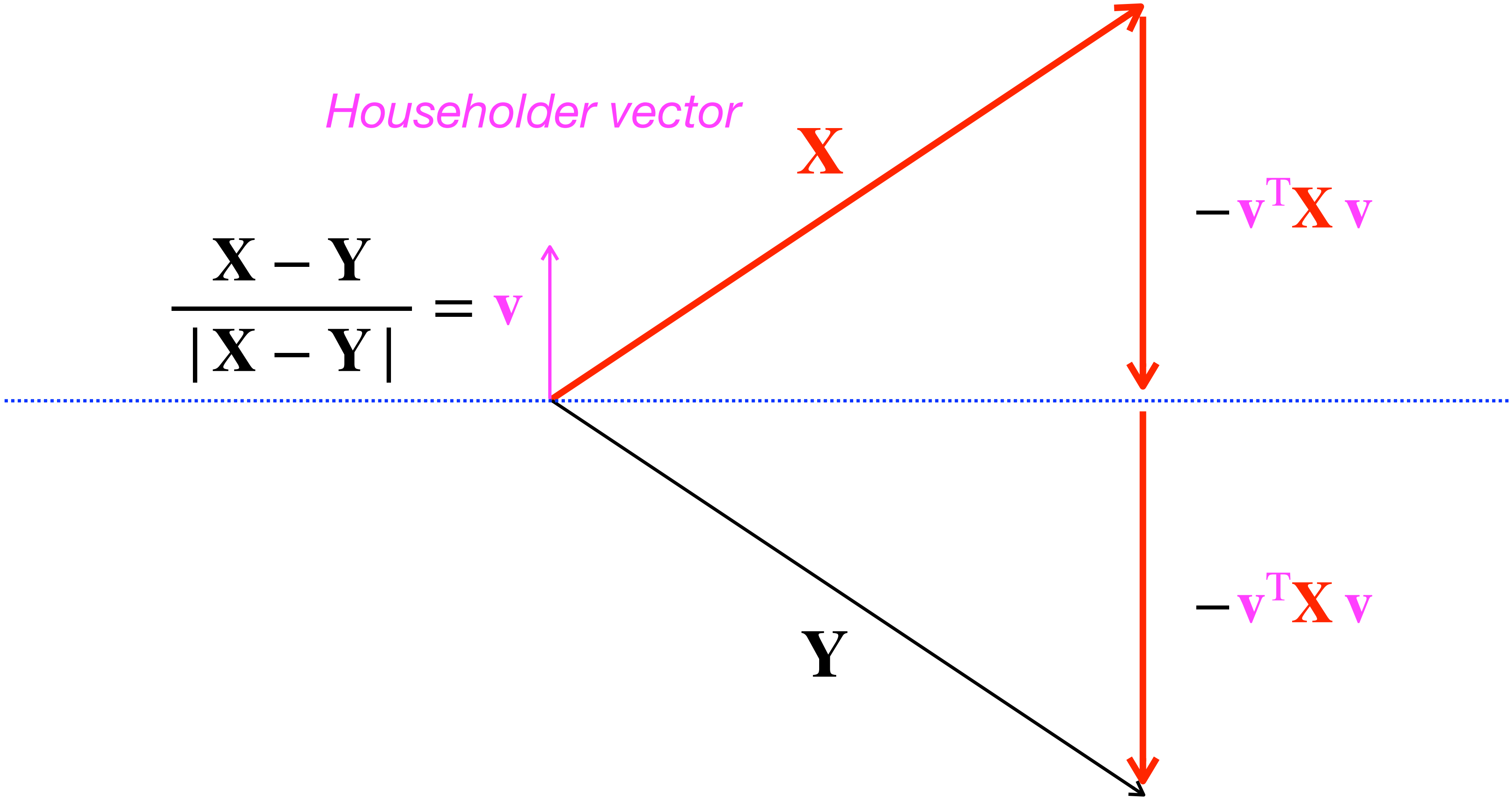
The Householder transformation



A. S. Householder, J. ACM 5, 339 (1958).
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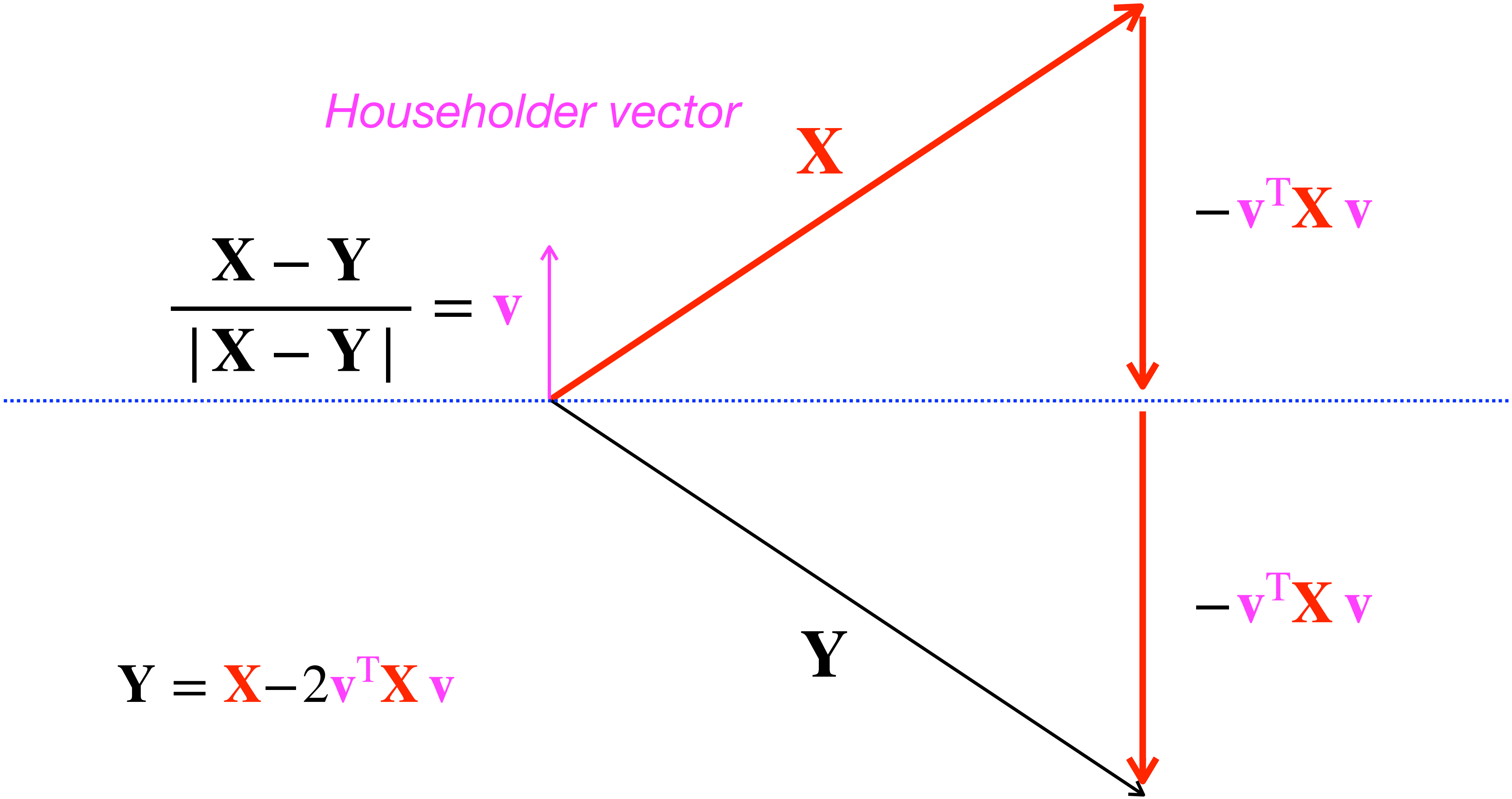
The Householder transformation



$$\frac{\mathbf{X}-\mathbf{Y}}{|\mathbf{X}-\mathbf{Y}|}=\mathbf{v}$$

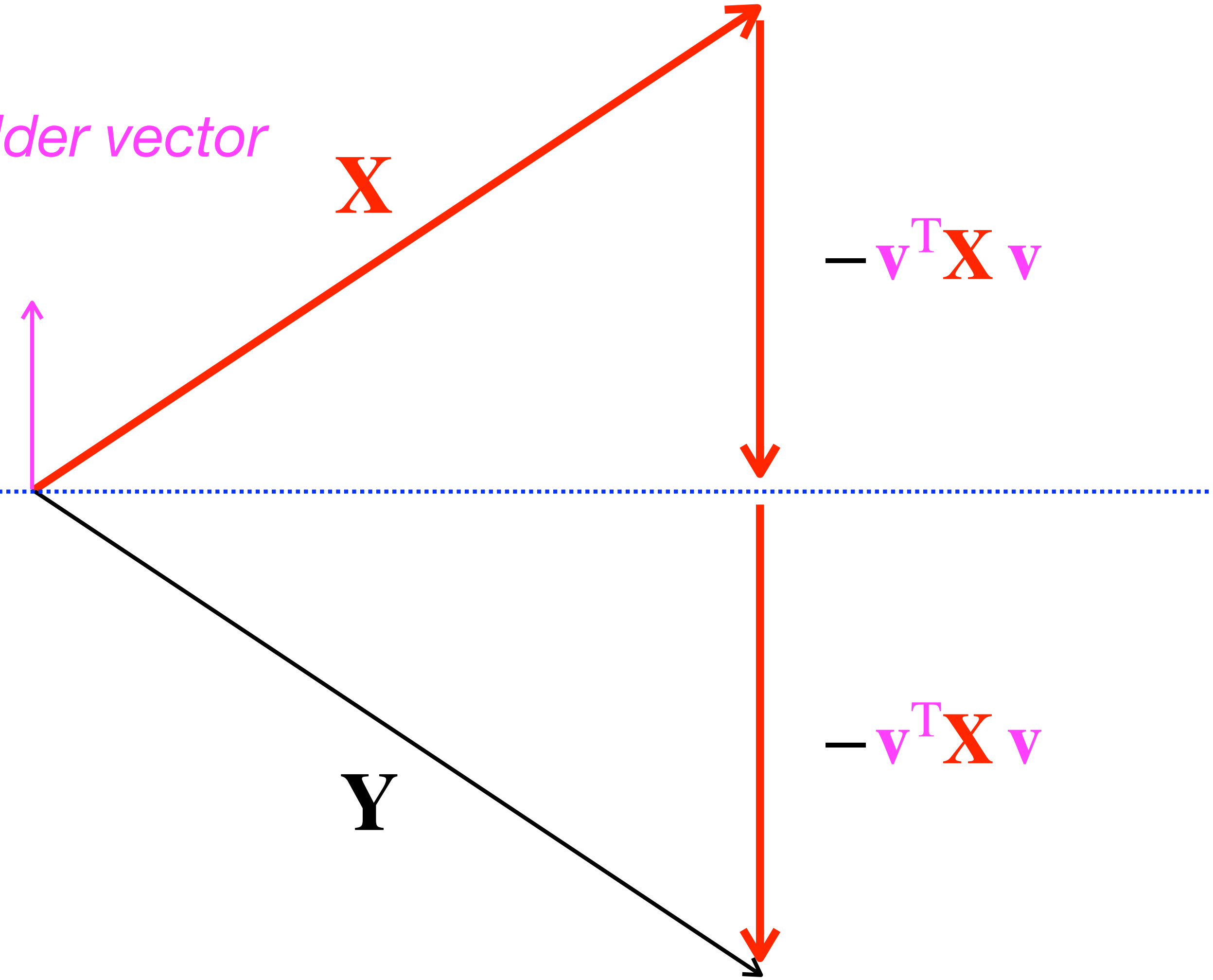
$$\mathbf{Y}=\mathbf{X}-2\mathbf{v}^T\mathbf{X}\,\mathbf{v}$$

A. S. Householder, J. ACM 5, 339 (1958).
*S. Sekaran, M. Tsuchiizu, M. Saubanère, and E. Fromager, Phys. Rev. B **104**, 035121 (2021).*

The Householder transformation

Householder vector

$$\frac{\mathbf{X}-\mathbf{Y}}{|\mathbf{X}-\mathbf{Y}|}=\mathbf{v}$$

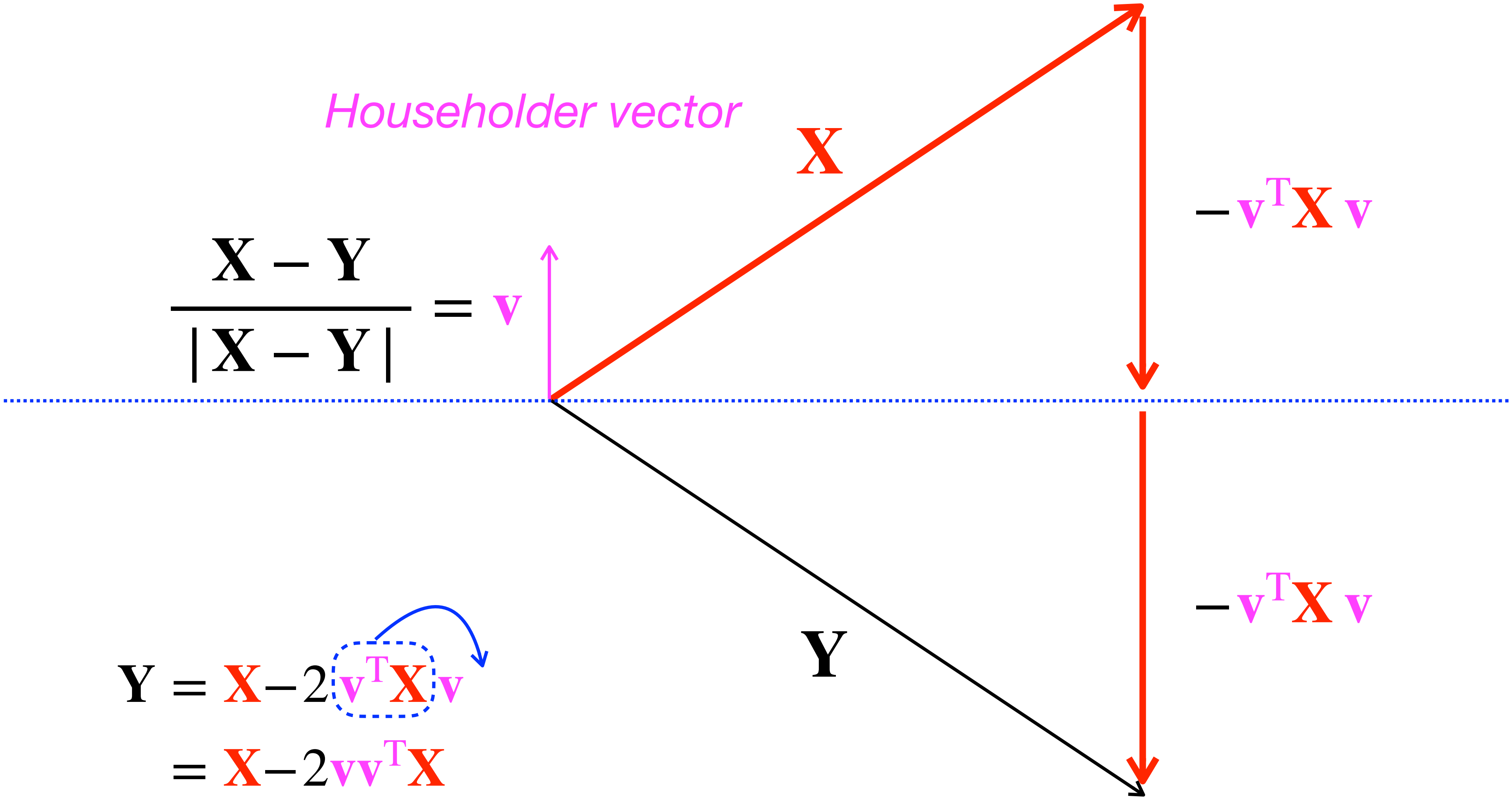


$$\mathbf{Y}=\mathbf{X}-2\,\mathbf{v}^{\mathrm{T}}\mathbf{X}\,\mathbf{v}$$
$$=\mathbf{X}-2\,\mathbf{v}\mathbf{v}^{\mathrm{T}}\mathbf{X}$$

A. S. Householder, J. ACM 5, 339 (1958).
*S. Sekaran, M. Tsuchiizu, M. Saubanère, and E. Fromager, Phys. Rev. B **104**, 035121 (2021).*

The Householder transformation

$$\frac{\mathbf{X}-\mathbf{Y}}{|\mathbf{X}-\mathbf{Y}|} = \mathbf{v}$$

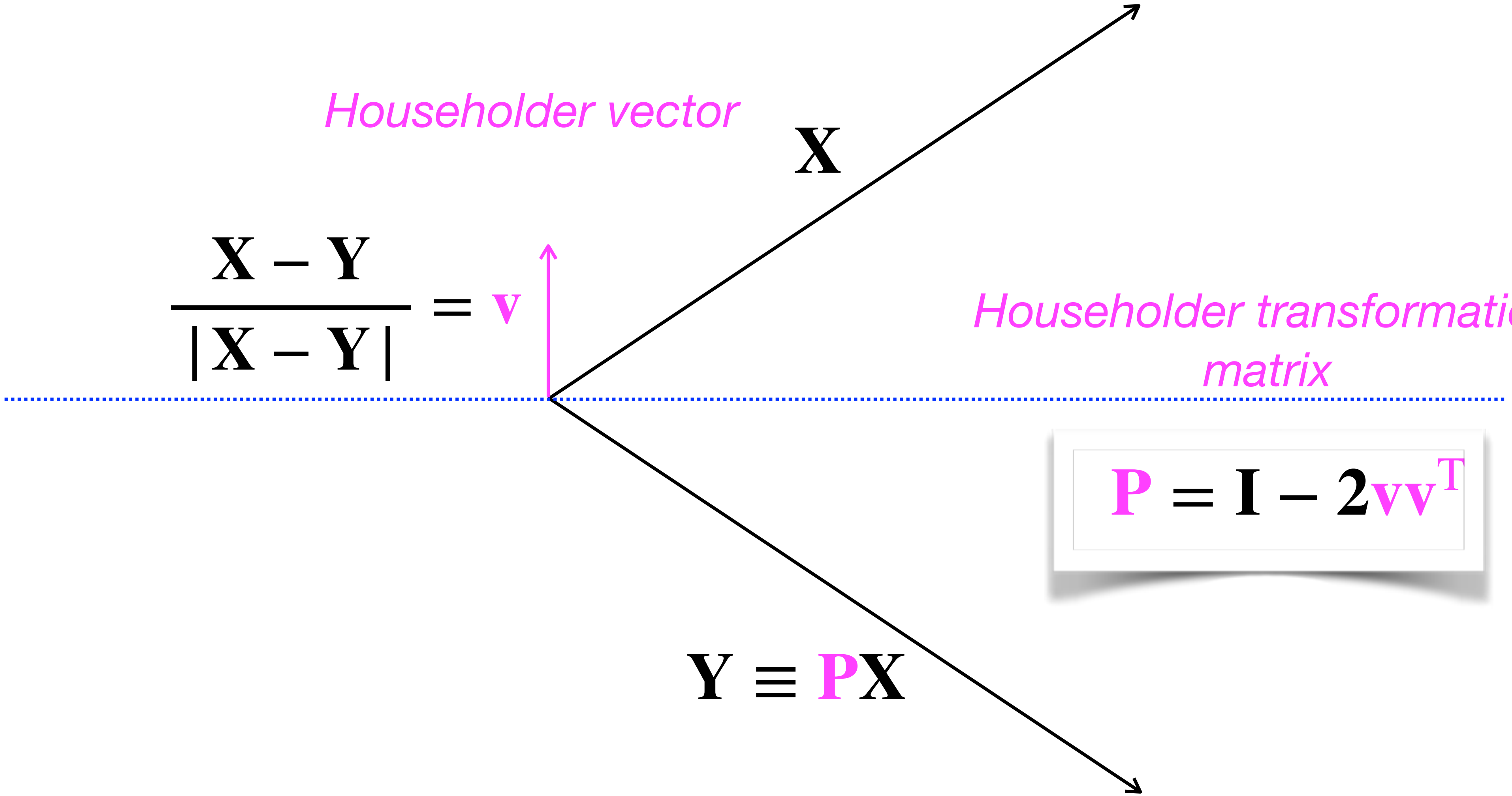


$$\begin{aligned}\mathbf{Y} &= \mathbf{X} - 2\,\mathbf{v}^{T}\mathbf{X}\,\mathbf{v}\\ &= \mathbf{X} - 2\,\mathbf{v}\mathbf{v}^{T}\mathbf{X}\\ &= \left(\mathbf{I} - 2\,\mathbf{v}\mathbf{v}^{T}\right)\mathbf{X}\end{aligned}$$

A. S. Householder, J. ACM 5, 339 (1958).

S. Sekaran, M. Tsuchiizu, M. Saubanère, and E. Fromager, Phys. Rev. B ***104****, 035121 (2021).*

The Householder transformation



$$\frac{\mathbf{X} - \mathbf{Y}}{|\mathbf{X} - \mathbf{Y}|} = \mathbf{v}$$

$$\mathbf{P} = \mathbf{I} - 2\mathbf{v}\mathbf{v}^{\mathrm{T}}$$

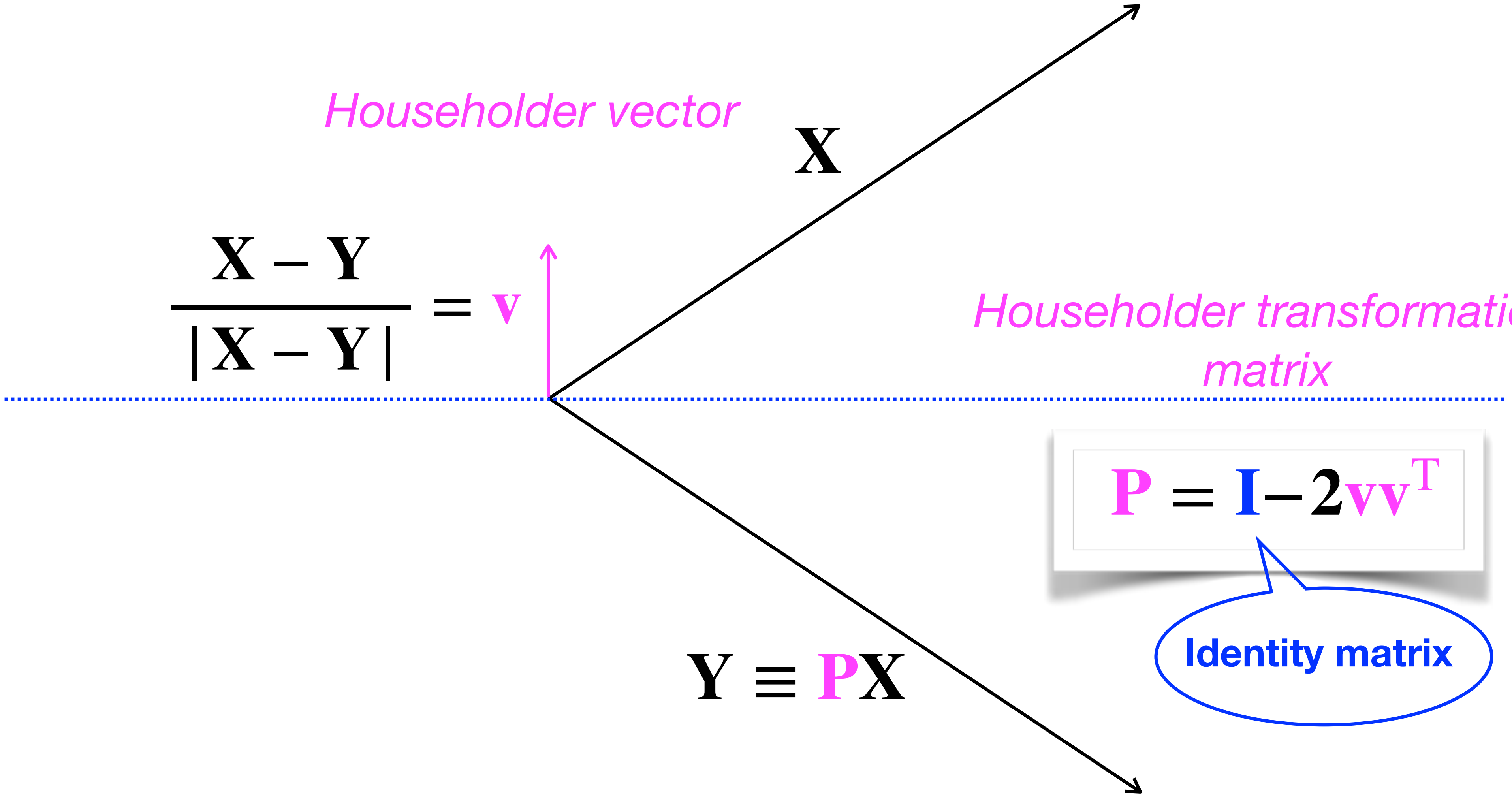
$$\mathbf{Y} \equiv \mathbf{P}\mathbf{X}$$

A. S. Householder, J. ACM 5, 339 (1958).
S. Sekaran, M. Tsuchiizu, M. Saubanère, and E. Fromager, Phys. Rev. B ***104****, 035121 (2021).*

The Householder transformation

Householder vector

$$\frac{\mathbf{X}-\mathbf{Y}}{|\mathbf{X}-\mathbf{Y}|}=\mathbf{v}$$



Householder transformation matrix

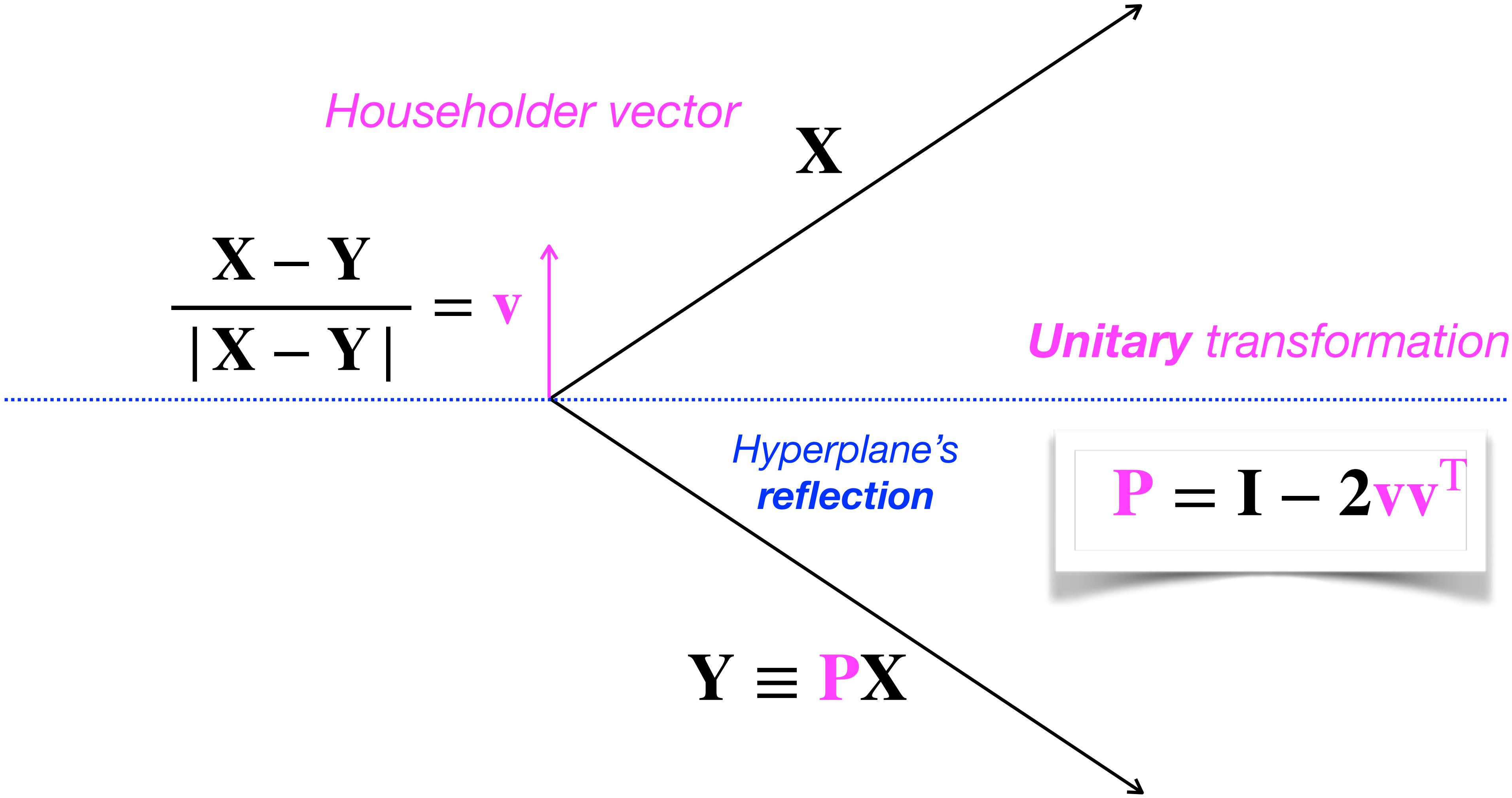
$$\mathbf{P}=\mathbf{I}-2\mathbf{v}\mathbf{v}^{\mathrm{T}}$$

Identity matrix

$$\mathbf{Y}\equiv\mathbf{P}\mathbf{X}$$

A. S. Householder, J. ACM 5, 339 (1958).
*S. Sekaran, M. Tsuchiizu, M. Saubanère, and E. Fromager, Phys. Rev. B **104**, 035121 (2021).*

The Householder transformation



A. S. Householder, J. ACM 5, 339 (1958).
*S. Sekaran, M. Tsuchiizu, M. Saubanère, and E. Fromager, Phys. Rev. B **104**, 035121 (2021).*

The Householder transformation

$$\mathbf{P}^{-1} = \mathbf{P}^{\mathrm{T}} = \mathbf{P}$$

Unitary *transformation*

$$\mathbf{P} = \mathbf{I} - 2\mathbf{v}\mathbf{v}^{\mathrm{T}}$$

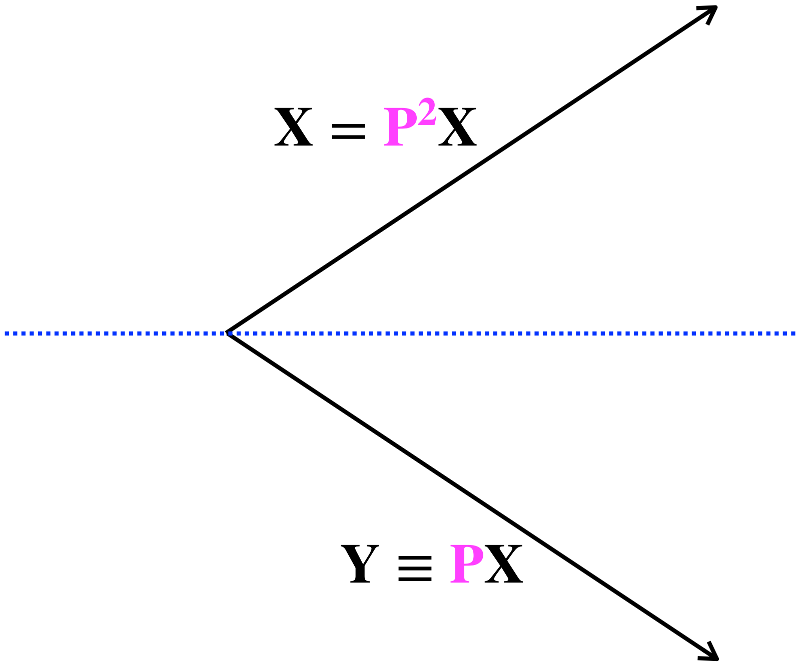
A. S. Householder, J. ACM 5, 339 (1958).
S. Sekaran, M. Tsuchiizu, M. Saubanère, and E. Fromager, Phys. Rev. B ***104****, 035121 (2021).*

The Householder transformation

$$\mathbf{P}^2 = \mathbf{I} \longleftarrow \mathbf{P}^{-1} = \mathbf{P}^{\mathrm{T}} = \mathbf{P}$$

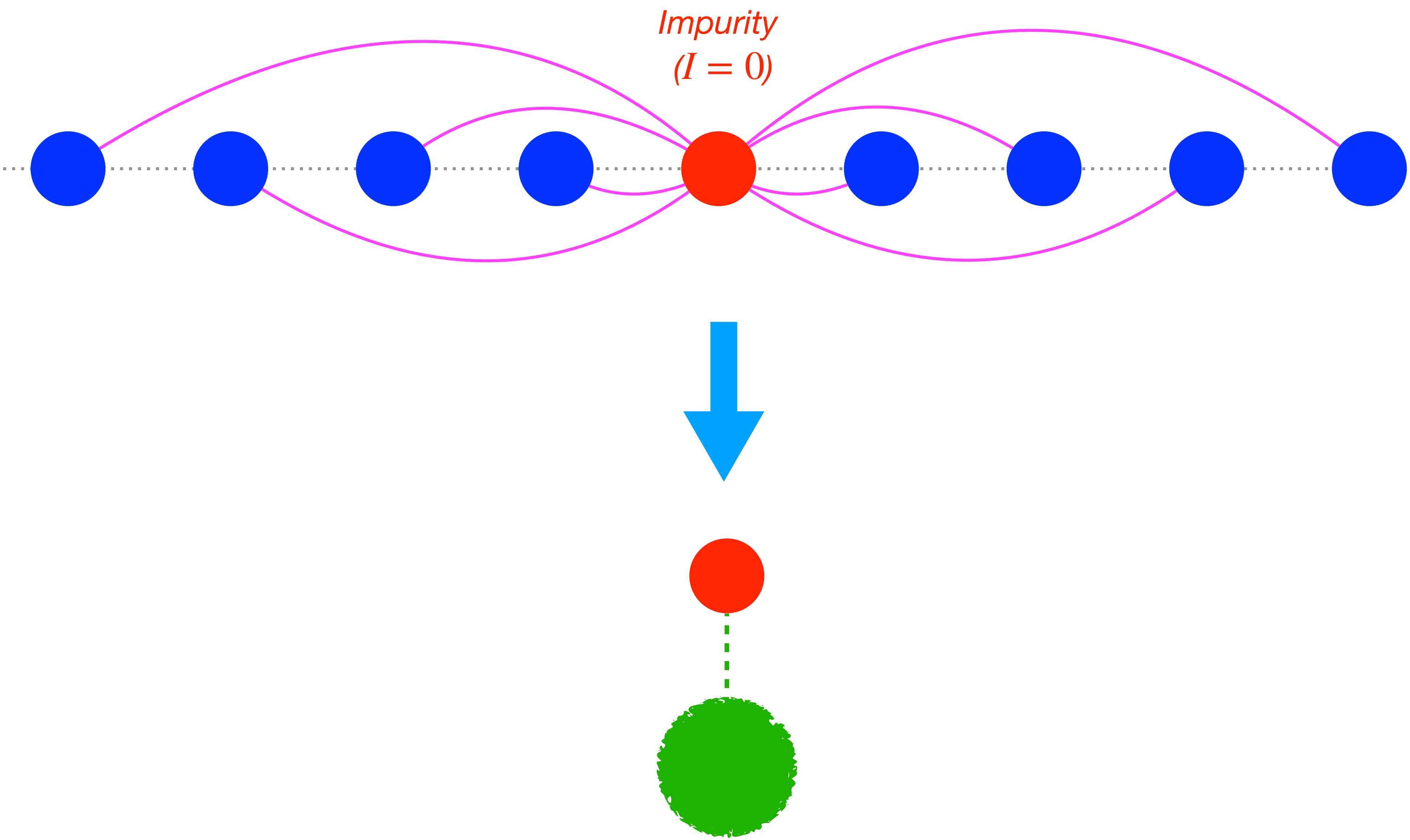
***Unitary** transformation*

$$\mathbf{P} = \mathbf{I} - 2\mathbf{v}\mathbf{v}^{\mathrm{T}}$$



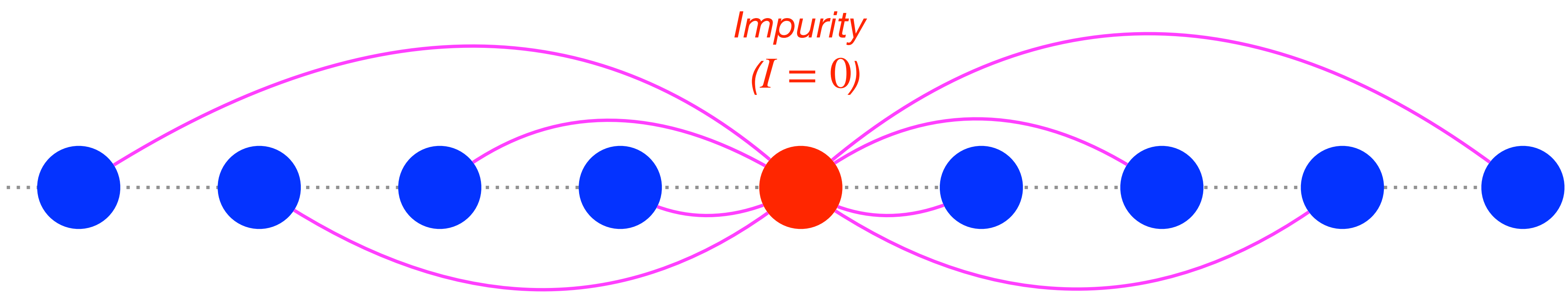
A. S. Householder, J. ACM 5, 339 (1958).
*S. Sekaran, M. Tsuchiizu, M. Saubanère, and E. Fromager, Phys. Rev. B **104**, 035121 (2021).*

Householder transformed density matrix embedding

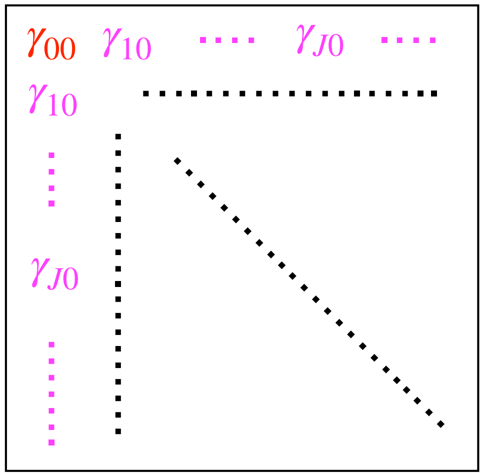


*G. Knizia and G. K.-L. Chan, Phys. Rev. Lett. **109**, 186404 (2012).*
*S. Sekaran, M. Tsuchiizu, M. Saubanère, and E. Fromager, Phys. Rev. B **104**, 035121 (2021).*

Householder transformed density matrix embedding



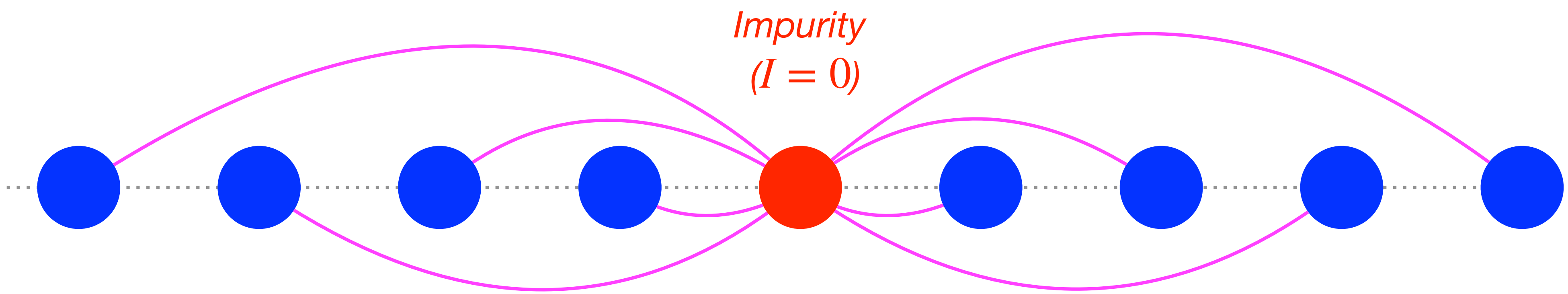
$$\gamma_{IJ} = \langle \hat{c}_I^\dagger \hat{c}_J \rangle \equiv$$

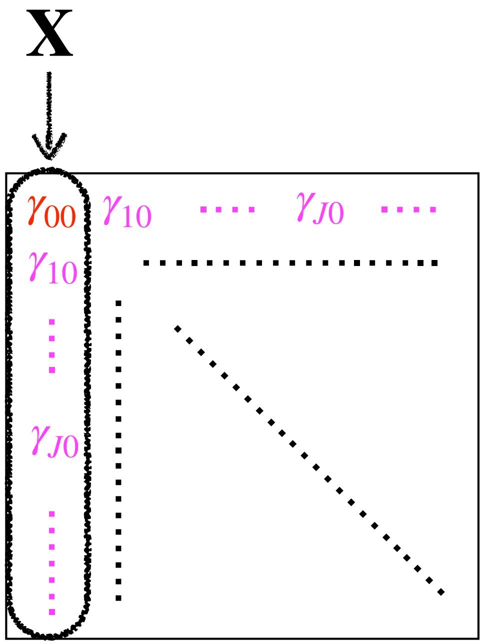


$$\gamma^{loc} \overset{notation}{=} \gamma$$

S. Sekaran, M. Tsuchiizu, M. Saubanère, and E. Fromager, Phys. Rev. B **104**, 035121 (2021).

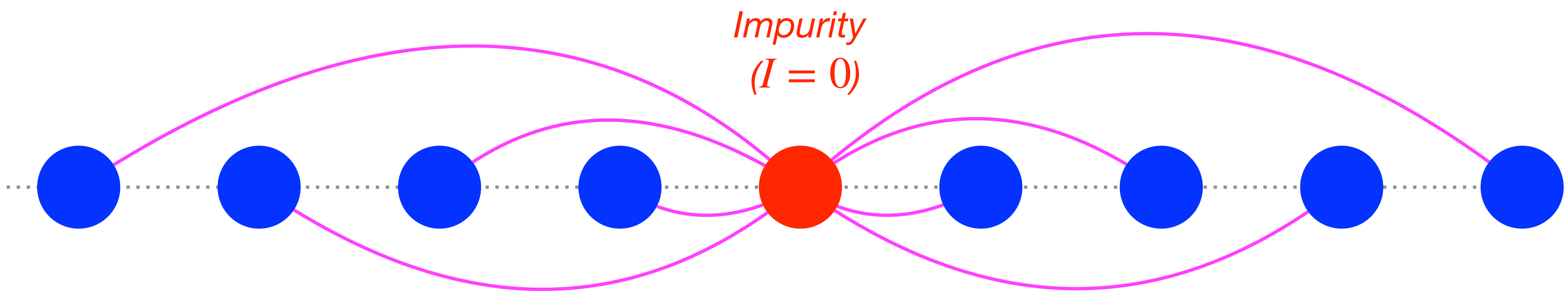
Householder transformed density matrix embedding

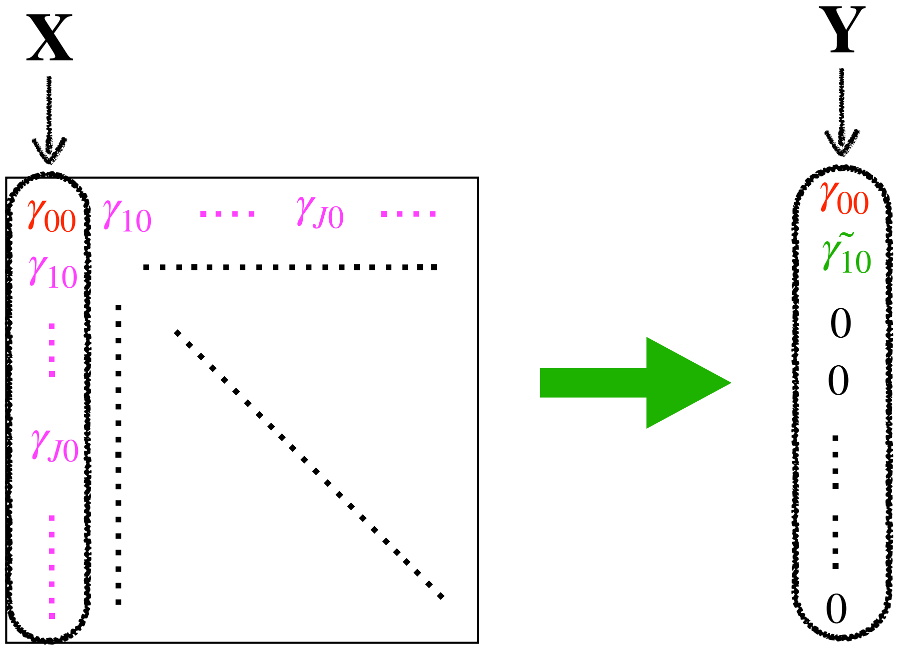




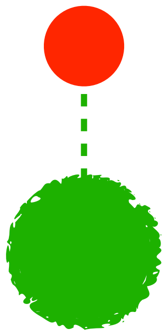
S. Sekaran, M. Tsuchiizu, M. Saubanère, and E. Fromager, Phys. Rev. B **104**, 035121 (2021).

Householder transformed density matrix embedding





“**X** *as we would like it to be*”



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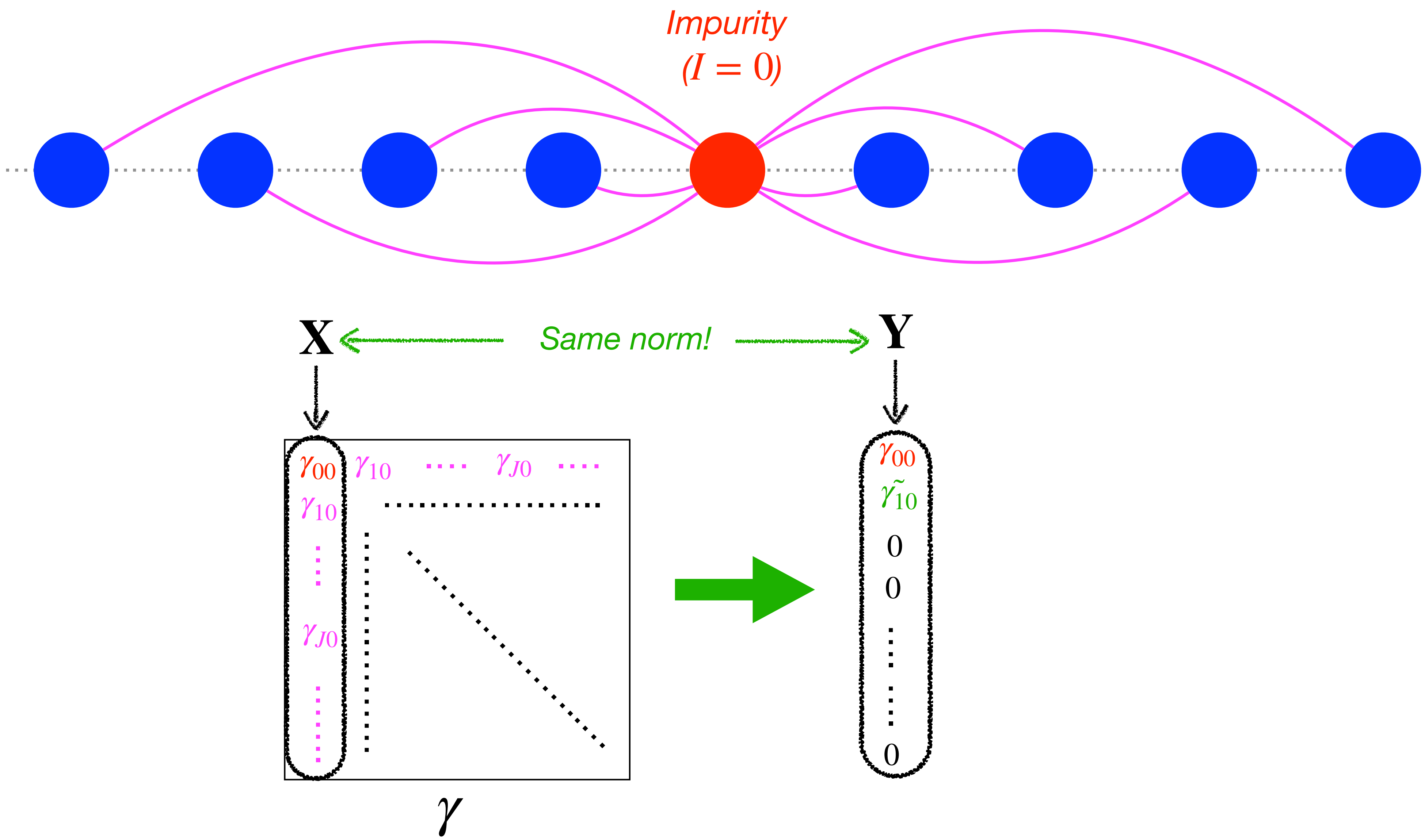
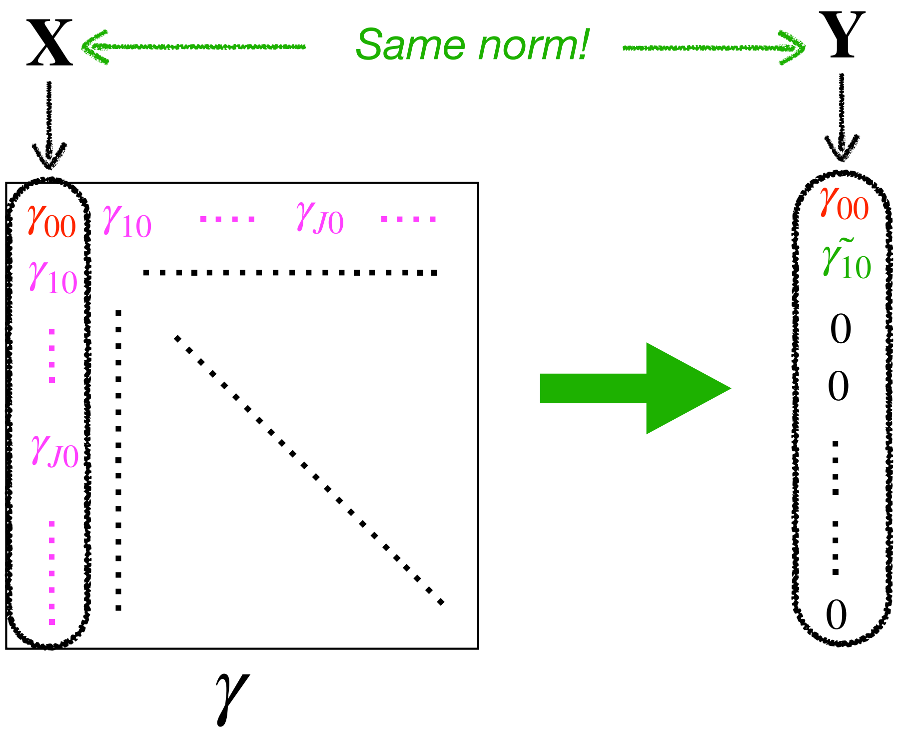
Householder transformed density matrix embedding



*S. Sekaran, M. Tsuchiizu, M. Saubanère, and E. Fromager, Phys. Rev. B **104**, 035121 (2021).*

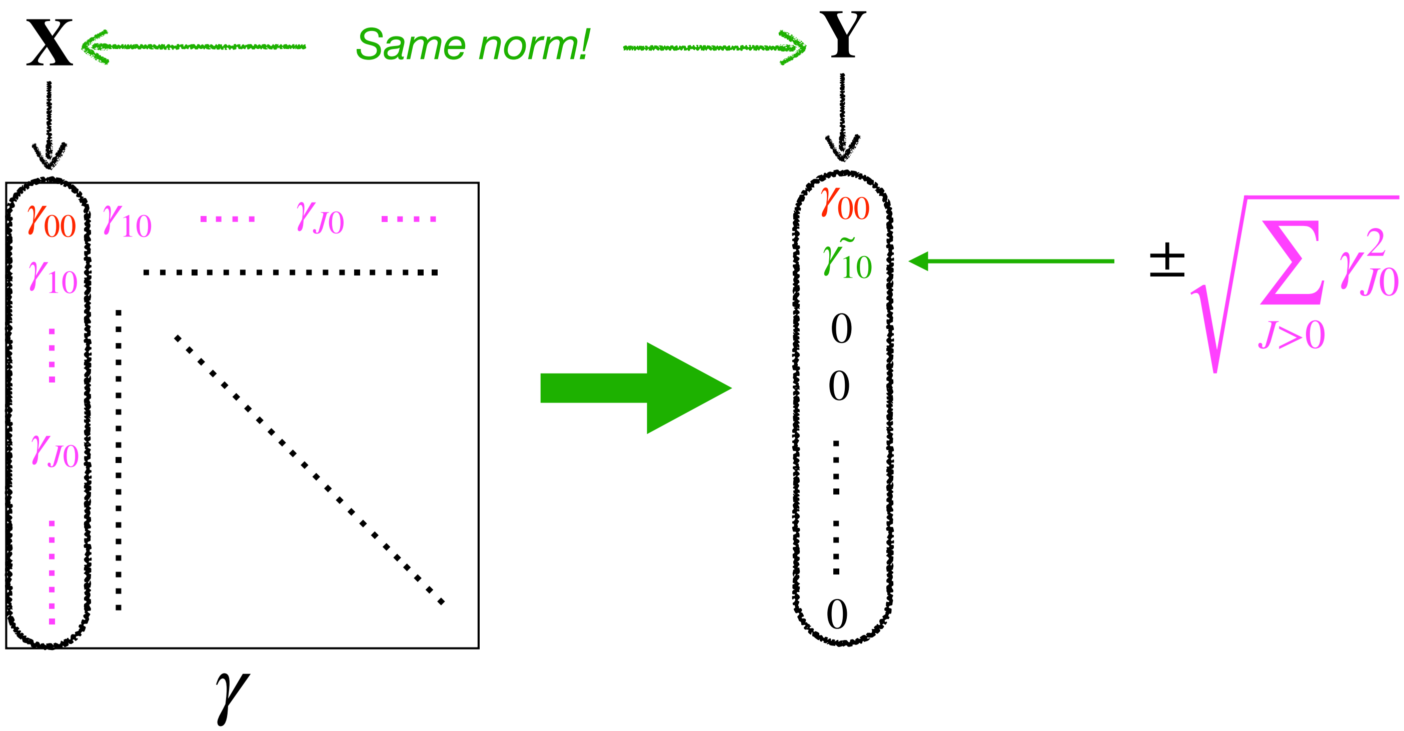
Householder transformed density matrix embedding

$$|\mathbf{X}|^2 - \gamma_{00}^2 = |\mathbf{Y}|^2 - \gamma_{00}^2 = \tilde{\gamma}_{10}^2 = \sum_{J>0} \gamma_{J0}^2$$



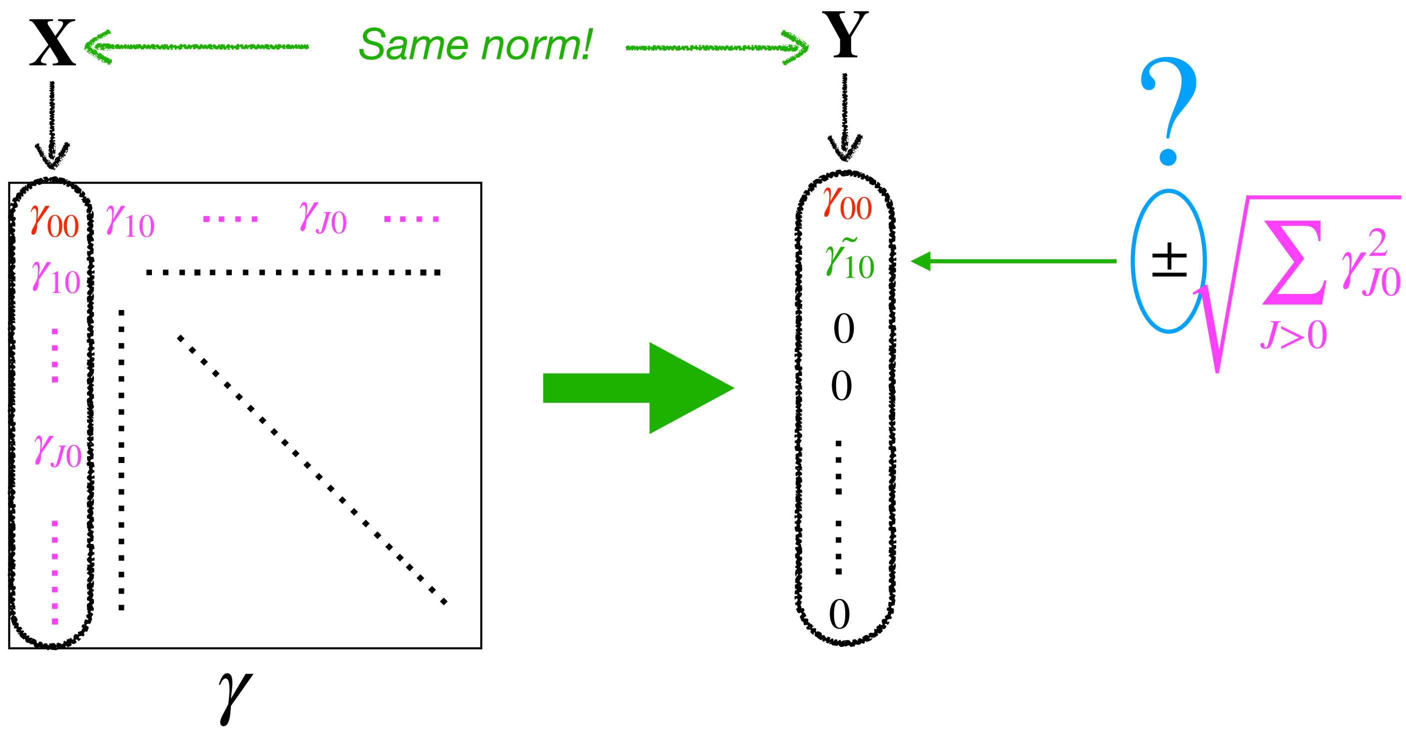
S. Sekaran, M. Tsuchiizu, M. Saubanère, and E. Fromager, Phys. Rev. B **104**, 035121 (2021).

Householder transformed density matrix embedding



S. Sekaran, M. Tsuchiizu, M. Saubanère, and E. Fromager, Phys. Rev. B **104**, 035121 (2021).

Householder transformed density matrix embedding

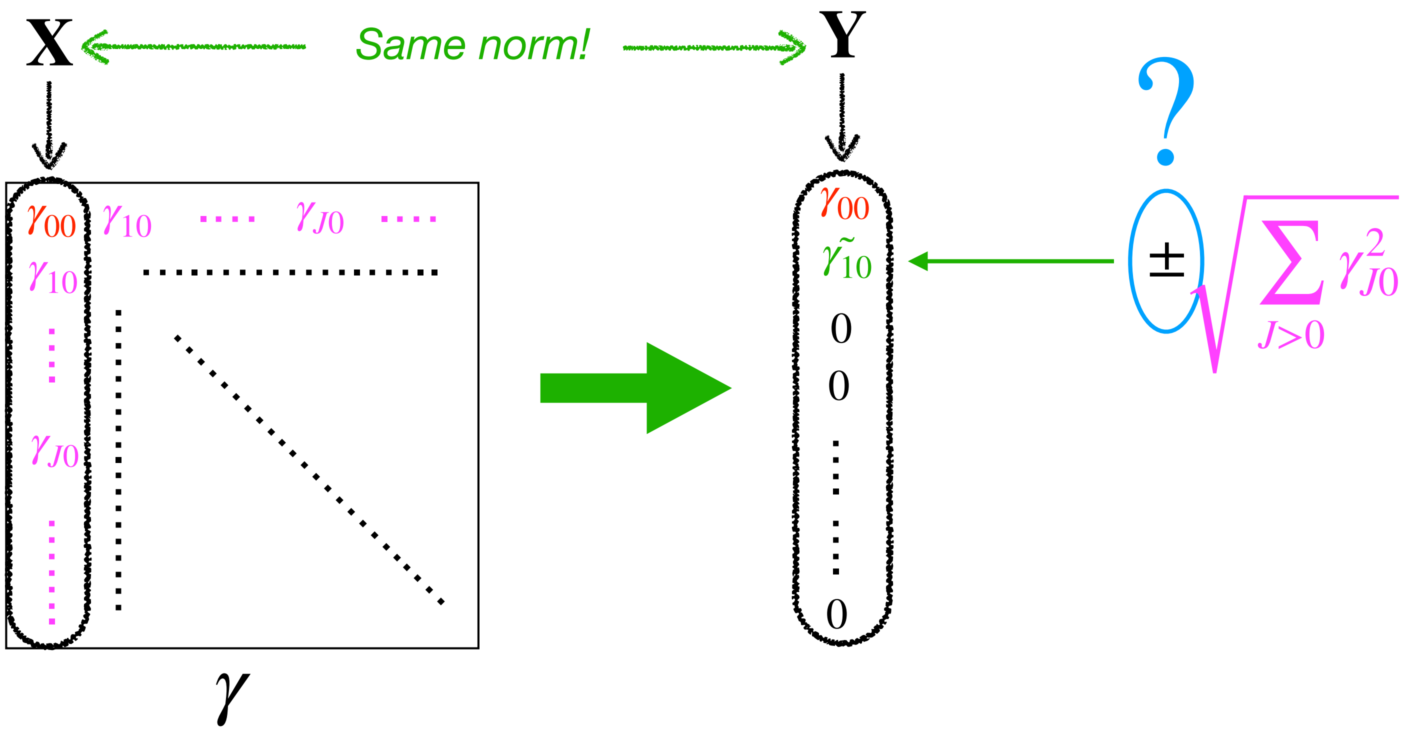


S. Sekaran, M. Tsuchiizu, M. Saubanère, and E. Fromager, Phys. Rev. B **104**, 035121 (2021).

Householder transformed density matrix embedding

$$\mathbf{v} = \frac{\mathbf{X} - \mathbf{Y}}{|\mathbf{X} - \mathbf{Y}|}$$ ← *Householder vector*

where $$|\mathbf{X} - \mathbf{Y}|^2 = 2\left(|\mathbf{Y}|^2 - \mathbf{X}^{\mathrm{T}}\mathbf{Y}\right) = 2\left(\tilde{\gamma}_{10}^2 - \gamma_{10}\tilde{\gamma}_{10}\right) = 2\tilde{\gamma}_{10}\left(\tilde{\gamma}_{10} - \gamma_{10}\right)$$



$$\tilde{\gamma}_{10} = \pm\sqrt{\sum_{J>0} \gamma_{J0}^2}$$

S. Sekaran, M. Tsuchiizu, M. Saubanère, and E. Fromager, Phys. Rev. B **104**, 035121 (2021).

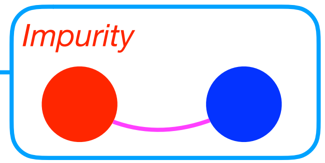
Householder transformed density matrix embedding

$$\mathbf{v} = \frac{\mathbf{X} - \mathbf{Y}}{|\mathbf{X} - \mathbf{Y}|}$$

Householder vector

where

$$|\mathbf{X} - \mathbf{Y}|^2 = \pm 2\sqrt{\sum_{J>0} \gamma_{J0}^2}\left(\pm\sqrt{\sum_{J>0} \gamma_{J0}^2} - \gamma_{10}\right)$$



If one single neighbour…

$$|\mathbf{X} - \mathbf{Y}|^2 = \pm 2|\gamma_{10}|\left(\pm|\gamma_{10}| - \gamma_{10}\right)$$

S. Sekaran, M. Tsuchiizu, M. Saubanère, and E. Fromager, Phys. Rev. B ***104****, 035121 (2021).*

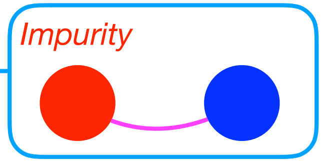
Householder transformed density matrix embedding

$$\mathbf{v} = \frac{\mathbf{X} - \mathbf{Y}}{|\mathbf{X} - \mathbf{Y}|}$$

Householder vector

where

$$|\mathbf{X} - \mathbf{Y}|^2 = \pm 2\sqrt{\sum_{J>0} \gamma_{J0}^2}\left(\pm\sqrt{\sum_{J>0} \gamma_{J0}^2} - \gamma_{10}\right)$$



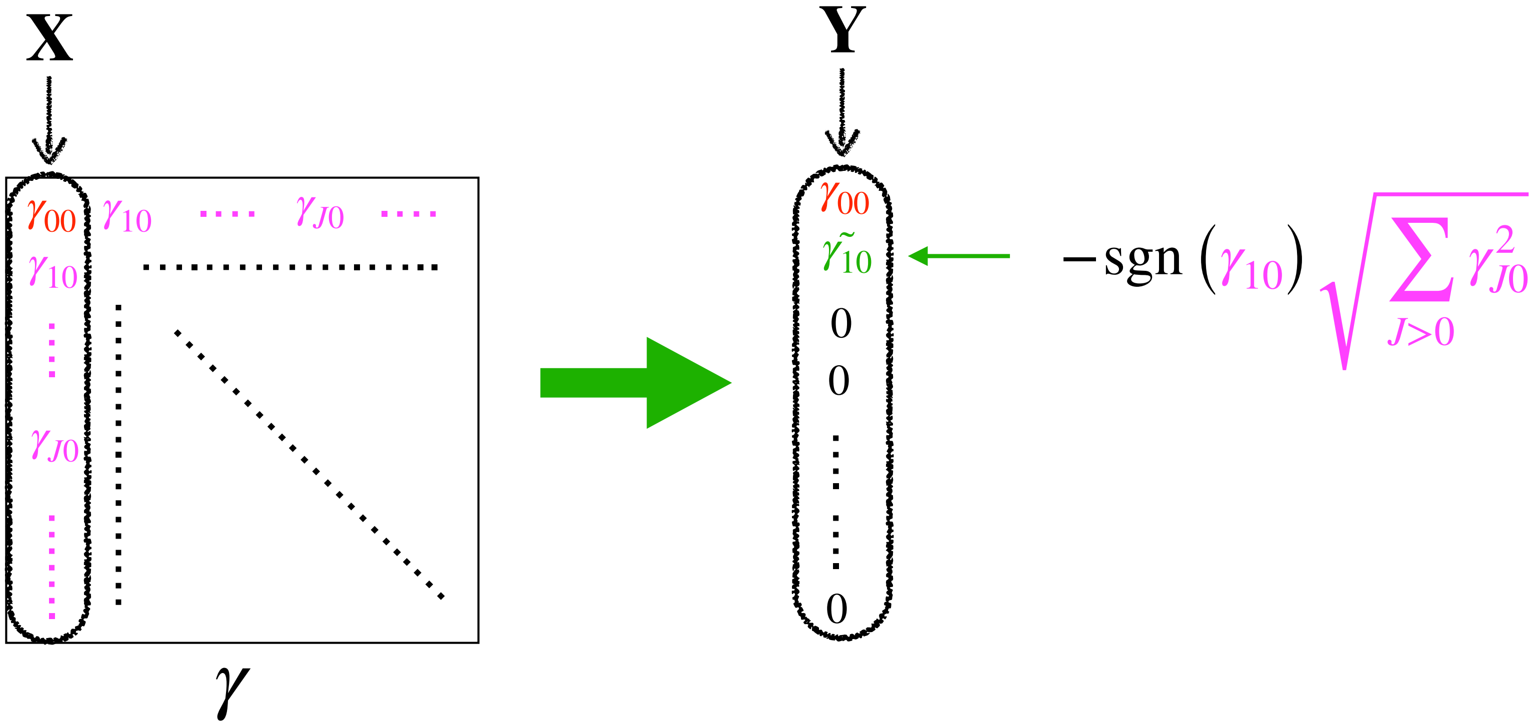
If one single neighbour…

$$|\mathbf{X} - \mathbf{Y}|^2 = \pm 2|\gamma_{10}|\left(\pm|\gamma_{10}| - \gamma_{10}\right)$$

choose $-\text{sgn}(\gamma_{10})$ ← $|\mathbf{X} - \mathbf{Y}| \neq 0$

S. Sekaran, M. Tsuchiizu, M. Saubanère, and E. Fromager, Phys. Rev. B **104**, 035121 (2021).

Householder transformed density matrix embedding



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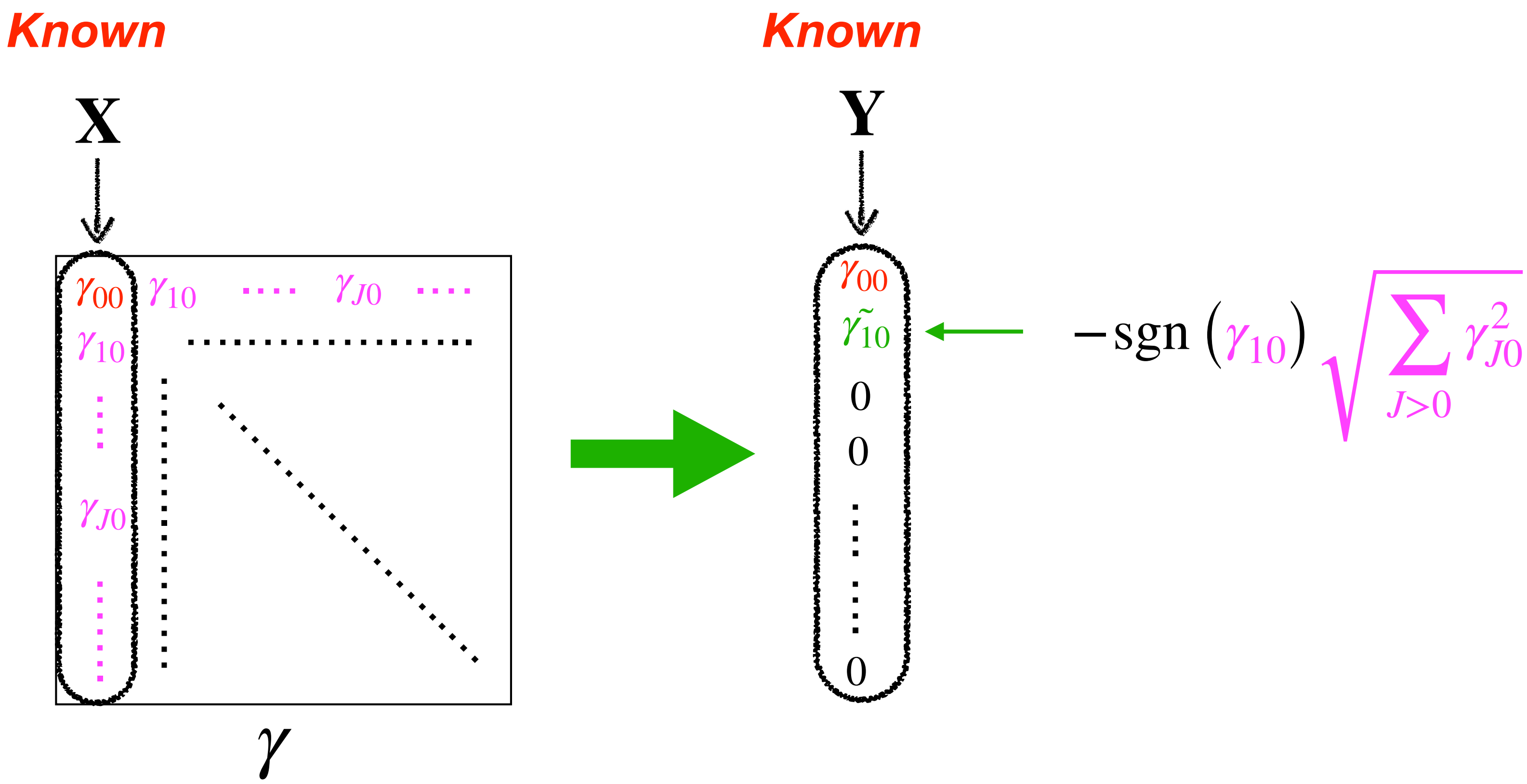
Householder transformed density matrix embedding



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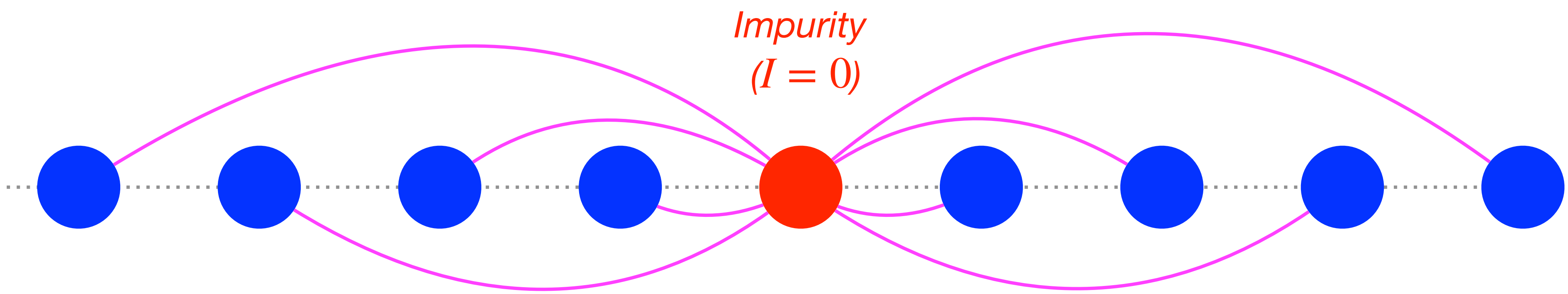
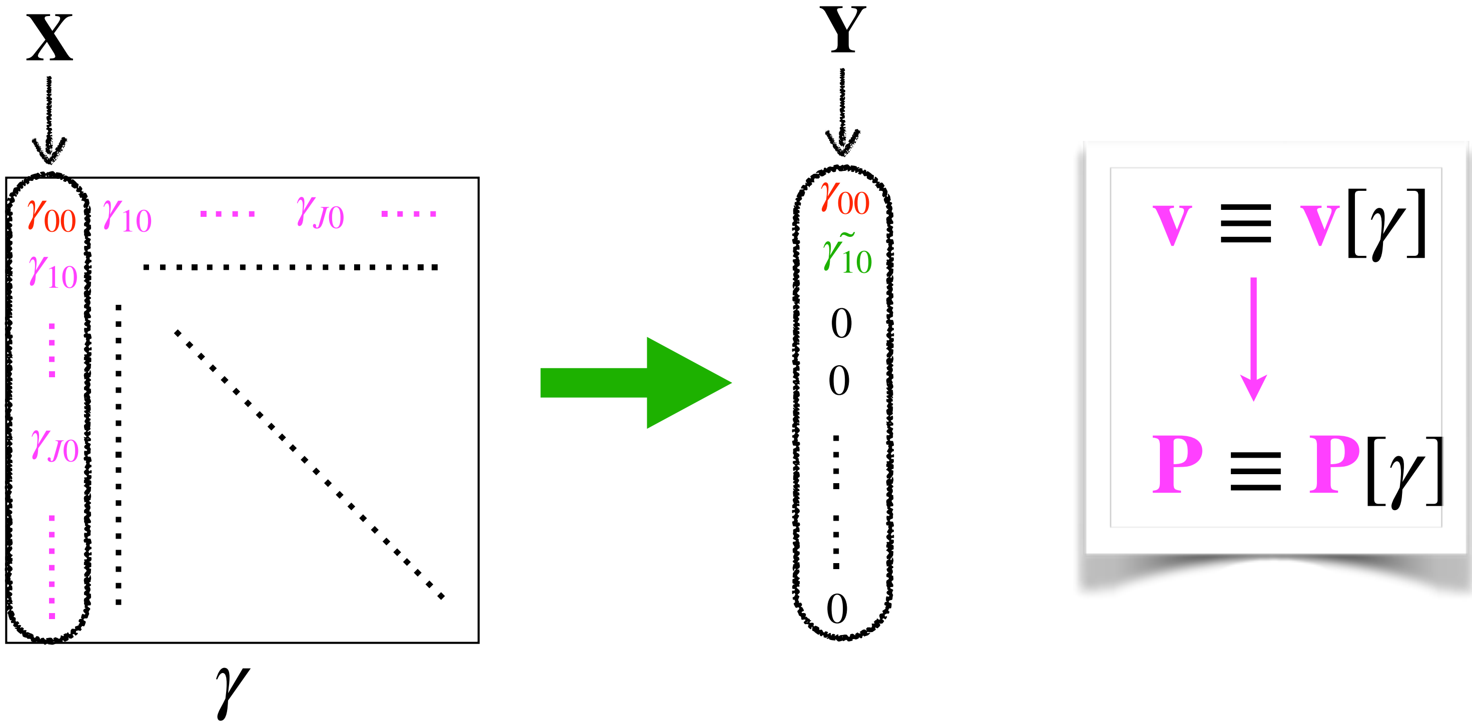
Householder transformed density matrix embedding





*The Householder transformation is an **explicit functional** of the density matrix!*

S. Sekaran, M. Tsuchiizu, M. Saubanère, and E. Fromager, Phys. Rev. B **104**, 035121 (2021).

Householder representation in second quantization

$$\mathbf{P} \equiv \mathbf{P}[\gamma] = \mathbf{I} - 2\mathbf{v}\mathbf{v}^{\mathrm{T}}$$

Unitary Householder transformation matrix

S. Sekaran, M. Tsuchiizu, M. Saubanère, and E. Fromager, Phys. Rev. B **104**, 035121 (2021).

Householder representation in second quantization

$$\mathbf{P} \equiv \mathbf{P}[\gamma] = \mathbf{I} - \mathbf{2vv}^{\mathrm{T}}$$

Unitary Householder transformation matrix

$$P_{IJ} = \delta_{IJ} - 2v_I v_J$$

*Householder transformation **matrix elements***

*S. Sekaran, M. Tsuchiizu, M. Saubanère, and E. Fromager, Phys. Rev. B **104**, 035121 (2021).*

Householder representation in second quantization

$$\mathbf{P} \equiv \mathbf{P}[\gamma] = \mathbf{I} - \mathbf{2}\mathbf{v}\mathbf{v}^{\mathrm{T}}$$

Unitary Householder transformation matrix

$$P_{IJ} = \delta_{IJ} - 2v_I v_J$$

Householder transformation ***matrix elements***

$$\hat{d}_I^\dagger = \sum_J P_{IJ}\, \hat{c}_J^\dagger$$

Creates delocalised ***Householder orbitals***

S. Sekaran, M. Tsuchiizu, M. Saubanère, and E. Fromager, Phys. Rev. B ***104****, 035121 (2021).*

Householder representation in second quantization

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$$\sum_I P_{KI}\hat{d}_I^\dagger = \sum_J \sum_I P_{KI}P_{IJ}\,\hat{c}_J^\dagger$$

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$$\sum_I P_{KI}\hat{d}_I^\dagger = \sum_J \sum_I P_{KI}P_{IJ}\, \hat{c}_J^\dagger = \sum_J \left[\mathbf{P}^2\right]_{KJ}\hat{c}_J^\dagger = \sum_J \delta_{KJ}\hat{c}_J^\dagger = \hat{c}_K^\dagger$$

*S. Sekaran, M. Tsuchiizu, M. Saubanère, and E. Fromager, Phys. Rev. B **104**, 035121 (2021).*

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$$\hat{d}_I^\dagger = \sum_J P_{IJ}\, \hat{c}_J^\dagger$$

From the ***localised*** *to the* ***Householder*** *representation*

$$\hat{c}_K^\dagger = \sum_I P_{KI}\, \hat{d}_I^\dagger$$

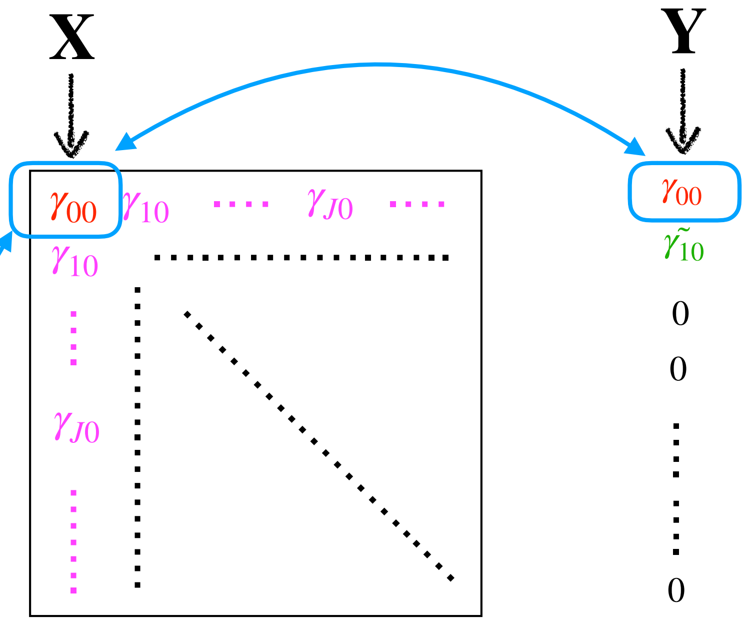
From the ***Householder*** *to the* ***localised*** *representation*

S. Sekaran, M. Tsuchiizu, M. Saubanère, and E. Fromager, Phys. Rev. B ***104****, 035121 (2021).*

Householder representation in second quantization

$$\mathbf{v} = \frac{\mathbf{X} - \mathbf{Y}}{|\mathbf{X} - \mathbf{Y}|}$$

Householder vector



$$\hat{d}_0^\dagger = \sum_J P_{0J}\, \hat{c}_J^\dagger \overset{v_0=0}{=} \hat{c}_0^\dagger$$

$$P_{0J} = \delta_{0J} - 2v_0 v_J$$

*The **impurity** is **invariant** under the transformation*

S. Sekaran, M. Tsuchiizu, M. Saubanère, and E. Fromager, Phys. Rev. B **104**, 035121 (2021).

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Creates delocalised ***Householder orbitals***

$$\hat{d}_0^\dagger = \hat{c}_0^\dagger$$

The ***impurity*** *is* ***invariant*** *under the transformation*

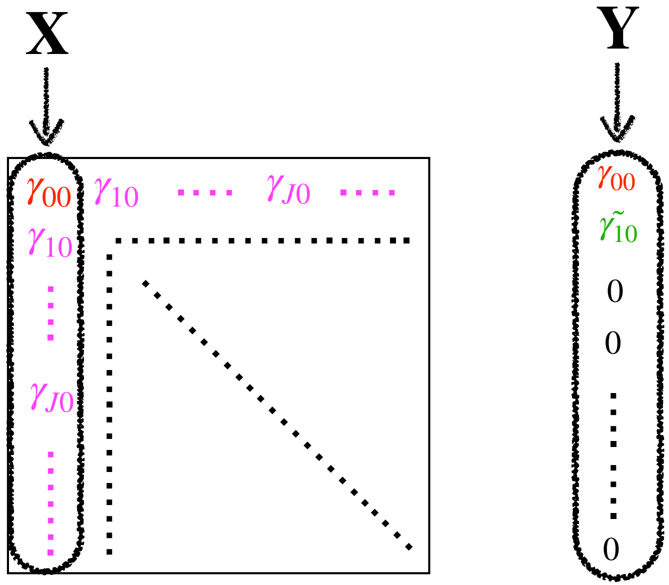
$$\hat{d}_1^\dagger |\mathrm{vac}\rangle = \sum_J P_{1J}\, |\chi_J\rangle \equiv |\varphi_{bath}\rangle$$

Will play the role of the ***bath spin-orbital***

S. Sekaran, M. Tsuchiizu, M. Saubanère, and E. Fromager, Phys. Rev. B ***104****, 035121 (2021).*

1RDM in the Householder representation

$$
\begin{aligned}
\tilde{\gamma}_{J0} &= \langle \Psi_0 | \hat{d}_J^\dagger \hat{d}_0 | \Psi_0 \rangle \\
&= \sum_I P_{JI} \langle \Psi_0 | \hat{c}_I^\dagger \hat{c}_0 | \Psi_0 \rangle \\
&= \sum_I P_{JI}\, \gamma_{I0} \\
&= \left[\mathbf{PX}\right]_J \\
&= \left[\mathbf{Y}\right]_J \\
&\overset{J\geq 1}{=} 0
\end{aligned}
$$



S. Sekaran, M. Tsuchiizu, M. Saubanère, and E. Fromager, Phys. Rev. B **104**, 035121 (2021).

1RDM in the Householder representation

$$\tilde{\gamma}_{J0} = \langle \Psi_0 | \hat{d}_J^\dagger \hat{d}_0 | \Psi_0 \rangle$$

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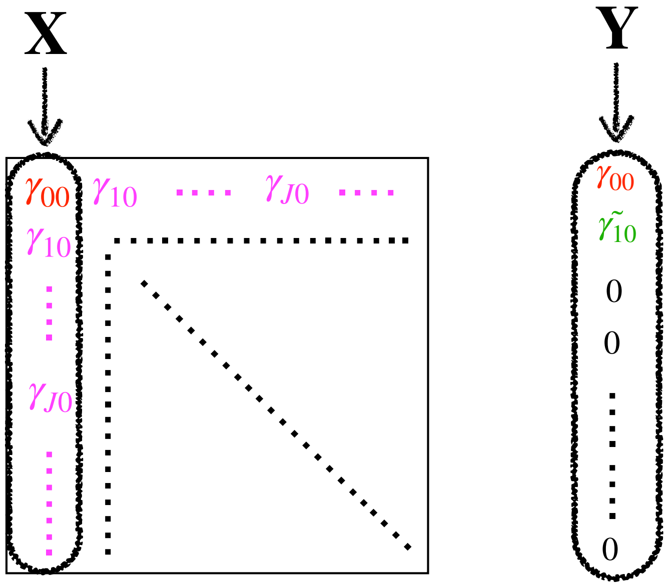

By construction, the impurity is entangled only with the bath

*S. Sekaran, M. Tsuchiizu, M. Saubanère, and E. Fromager, Phys. Rev. B **104**, 035121 (2021).*

1RDM in the Householder representation

Theorem: *As the electrons are **non-interacting**, the bath turns out to be **entangled only with the impurity***

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Proof

$$\tilde{\gamma} = \tilde{\gamma}^2$$

Idempotency property

*S. Sekaran, M. Tsuchiizu, M. Saubanère, and E. Fromager, Phys. Rev. B **104**, 035121 (2021).*

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Idempotency property

$$\tilde{\gamma}_{J0} = \langle \Psi_0 | \hat{d}_J^\dagger \hat{d}_0 | \Psi_0 \rangle = \left[\tilde{\gamma}^2\right]_{J0} = \sum_K \tilde{\gamma}_{JK} \tilde{\gamma}_{K0}$$

$$= \tilde{\gamma}_{J0} \tilde{\gamma}_{00} + \tilde{\gamma}_{J1} \tilde{\gamma}_{10} + \sum_{K>1} \tilde{\gamma}_{JK} \times 0$$

S. Sekaran, M. Tsuchiizu, M. Saubanère, and E. Fromager, Phys. Rev. B **104**, 035121 (2021).

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$$\tilde{\gamma}_{J0} = \tilde{\gamma}_{J0}\tilde{\gamma}_{00} + \tilde{\gamma}_{J1}\tilde{\gamma}_{10}$$

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$$\tilde{\gamma}_{J1} = \frac{\tilde{\gamma}_{J0}\left(1 - \tilde{\gamma}_{00}\right)}{\tilde{\gamma}_{10}}$$

$J > 1$ → $\tilde{\gamma}_{J0} = 0$

No entanglement between the impurity and the orbitals other than the bath

$$\tilde{\gamma}_{J1} = 0$$

S. Sekaran, M. Tsuchiizu, M. Saubanère, and E. Fromager, Phys. Rev. B **104**, 035121 (2021).

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No entanglement between the bath and the orbitals other than the impurity!

$$\tilde{\gamma}_{J1} = \langle \Psi_0 | \hat{d}_J^{\dagger} \hat{d}_1 | \Psi_0 \rangle = 0$$

S. Sekaran, M. Tsuchiizu, M. Saubanère, and E. Fromager, Phys. Rev. B **104**, 035121 (2021).

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$$\tilde{\gamma}_{J1} = \frac{\tilde{\gamma}_{J0}\left(1 - \tilde{\gamma}_{00}\right)}{\tilde{\gamma}_{10}}$$

$J = 1$

$$\tilde{\gamma}_{11} + \tilde{\gamma}_{00} = 1$$

S. Sekaran, M. Tsuchiizu, M. Saubanère, and E. Fromager, Phys. Rev. B **104**, 035121 (2021).

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*The "impurity+bath" cluster contains exactly **one electron (per spin)***

*S. Sekaran, M. Tsuchiizu, M. Saubanère, and E. Fromager, Phys. Rev. B **104**, 035121 (2021).*

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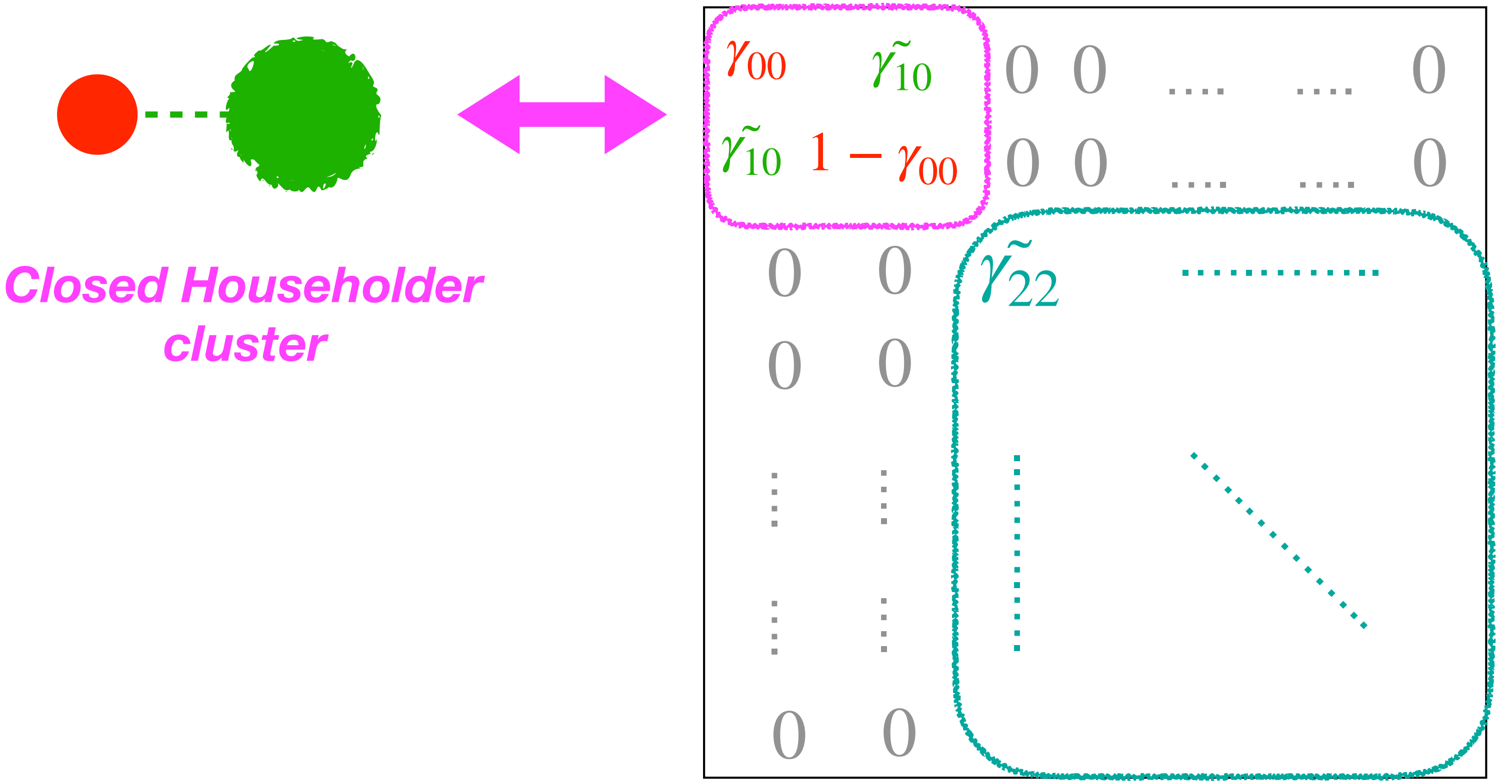
*The "impurity+bath" cluster contains exactly **one electron (per spin)***

*The cluster is a **closed quantum system** that can be described with a two-electron wave function* $\Psi^{\mathscr{C}}$

S. Sekaran, M. Tsuchiizu, M. Saubanère, and E. Fromager, Phys. Rev. B **104**, 035121 (2021).

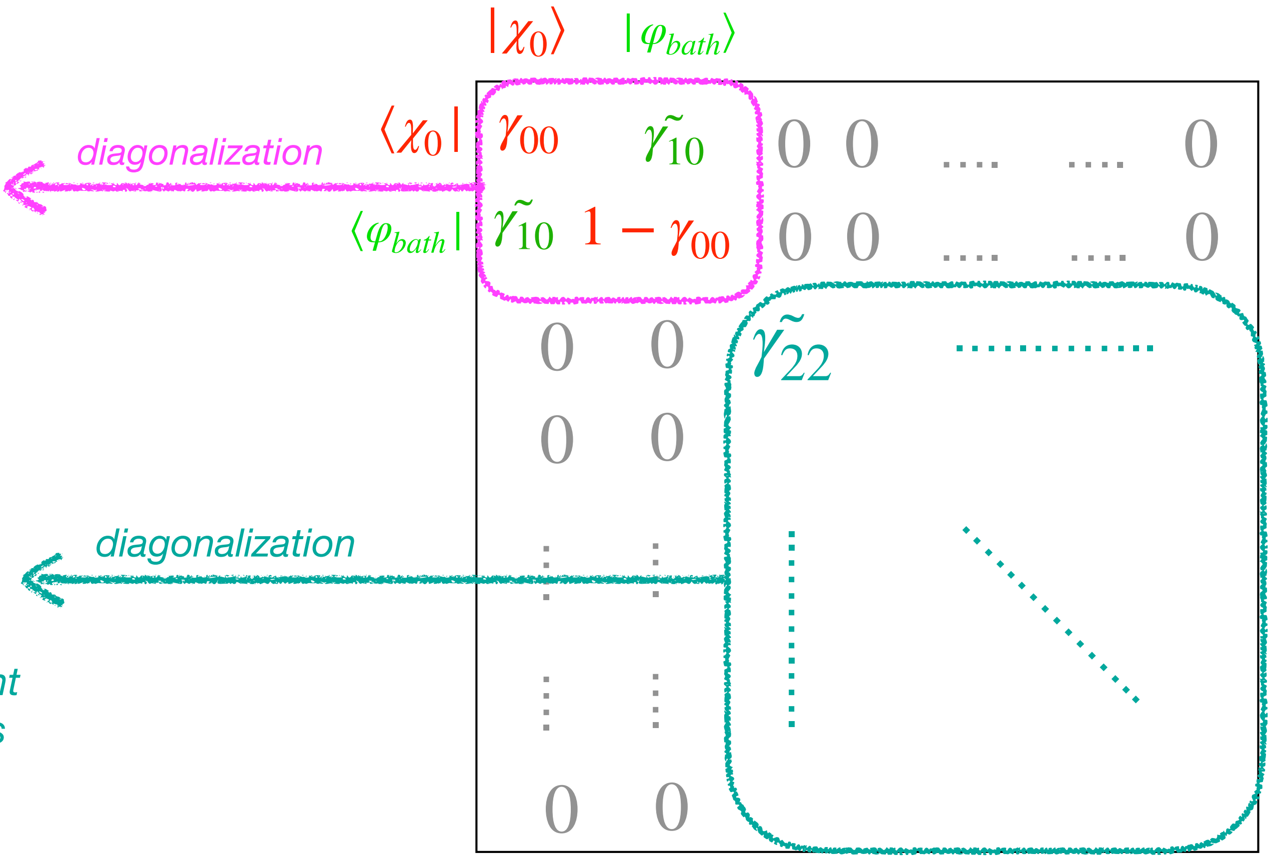
1RDM in the Householder representation



*S. Sekaran, M. Tsuchiizu, M. Saubanère, and E. Fromager, Phys. Rev. B **104**, 035121 (2021).*

1RDM in the Householder representation

$$\varphi^{\mathscr{C}}(\mathbf{x}) = C_0\chi_0(\mathbf{x}) + C_{bath}\varphi_{bath}(\mathbf{x})$$



$$\Phi_{core}$$

Slater determinant of "core" orbitals

*S. Sekaran, M. Tsuchiizu, M. Saubanère, and E. Fromager, Phys. Rev. B **104**, 035121 (2021).*

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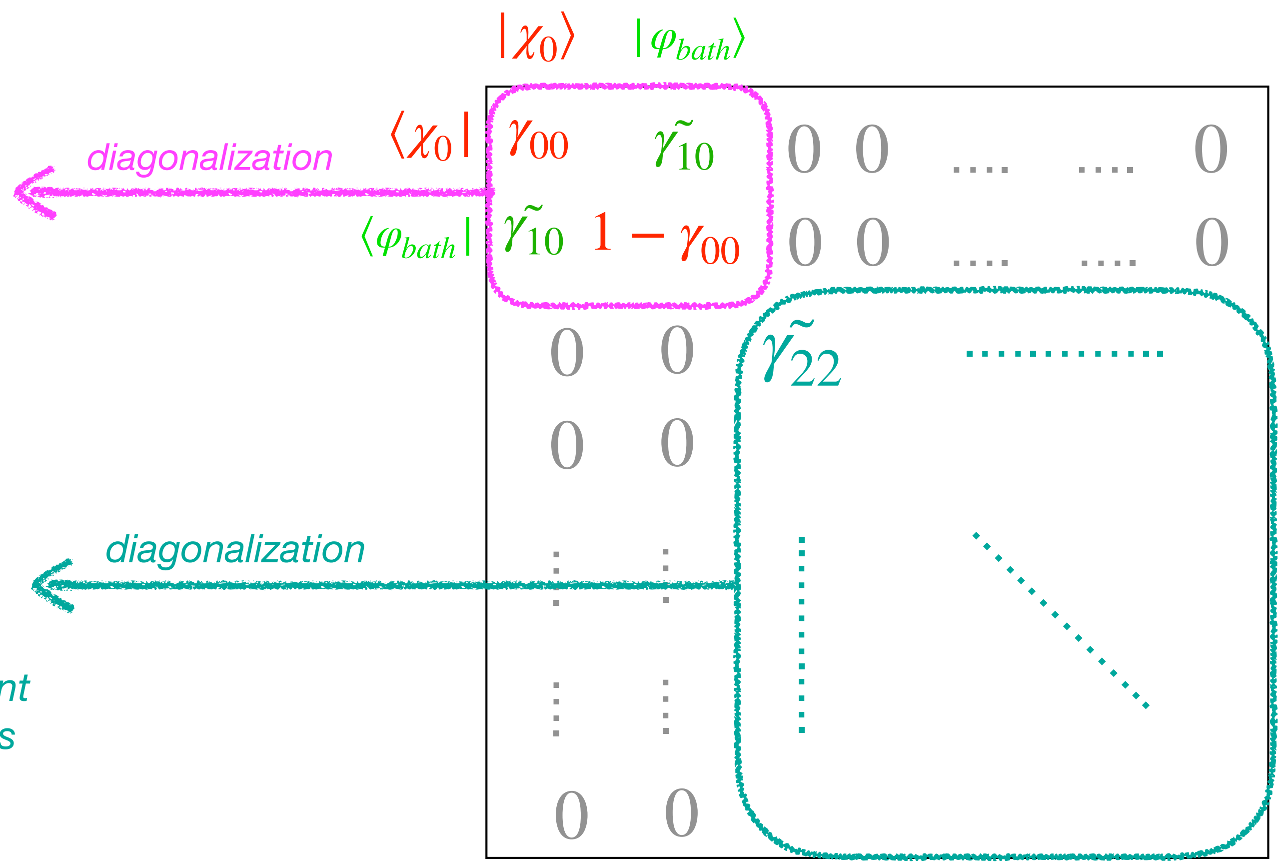

Φ_{core}

Slater determinant
of “core” orbitals

$$\Psi_0 \equiv \left(\varphi^{\mathscr{C}}\right)^2 \Phi_{core}$$

S. Sekaran, M. Tsuchiizu, M. Saubanère, and E. Fromager, Phys. Rev. B **104**, 035121 (2021).

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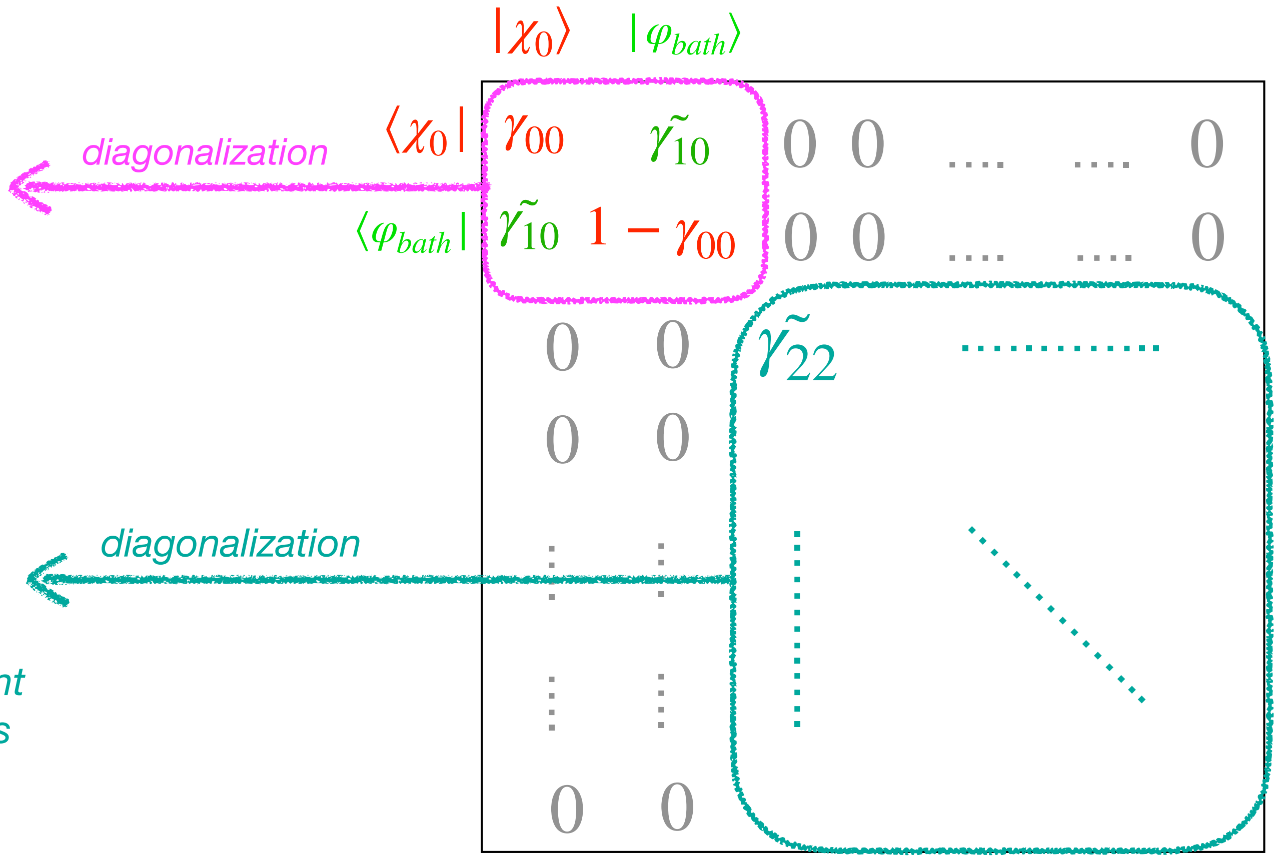

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*After **diagonalising each block**, we realise that, among all the occupied molecular orbitals in the full-system Slater determinant Ψ_0, a **single one** $\varphi^{\mathscr{C}}$ has a **nonzero overlap with the impurity**.*

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*The **Schmidt decomposition** of Ψ_0*
(which is obtained from the singular value decomposition of a CI coefficients matrix)
*leads to the **exact same result**[1,2].*

[1]*S. Sekaran, M. Tsuchiizu, M. Saubanère, and E. Fromager, Phys. Rev. B **104**, 035121 (2021).*
[2]*S. Wouters, C. A. Jiménez-Hoyos, Q. Sun, and G. K.-L. Chan, J. Chem. Theory Comput. **12**, 2706 (2016).*

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Important conclusion:

For non-interacting (or mean-field-like) electrons,
*the **Householder transformation** is **equivalent** to (but simpler than)*
*the **Schmidt decomposition**, which is central in DMET.*

[1] *S. Sekaran, M. Tsuchiizu, M. Saubanère, and E. Fromager, Phys. Rev. B **104**, 035121 (2021).*
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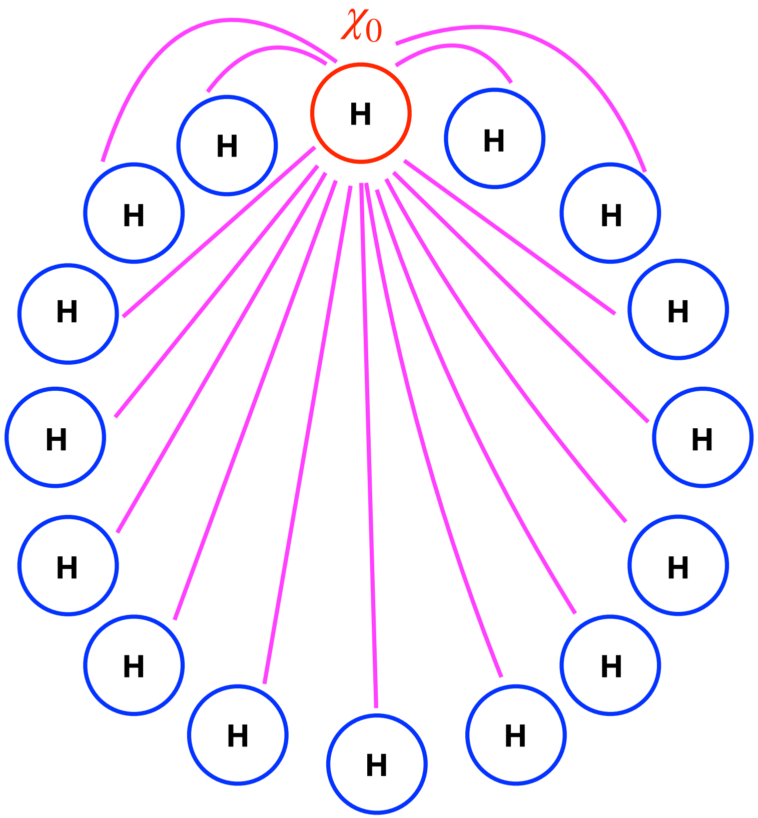
Energy evaluation by fragmentation

$$\hat{H} = \sum_{IJ} \overline{h}_{IJ} \, \hat{c}_I^\dagger \hat{c}_J$$

Localised representation

Energy evaluation by fragmentation



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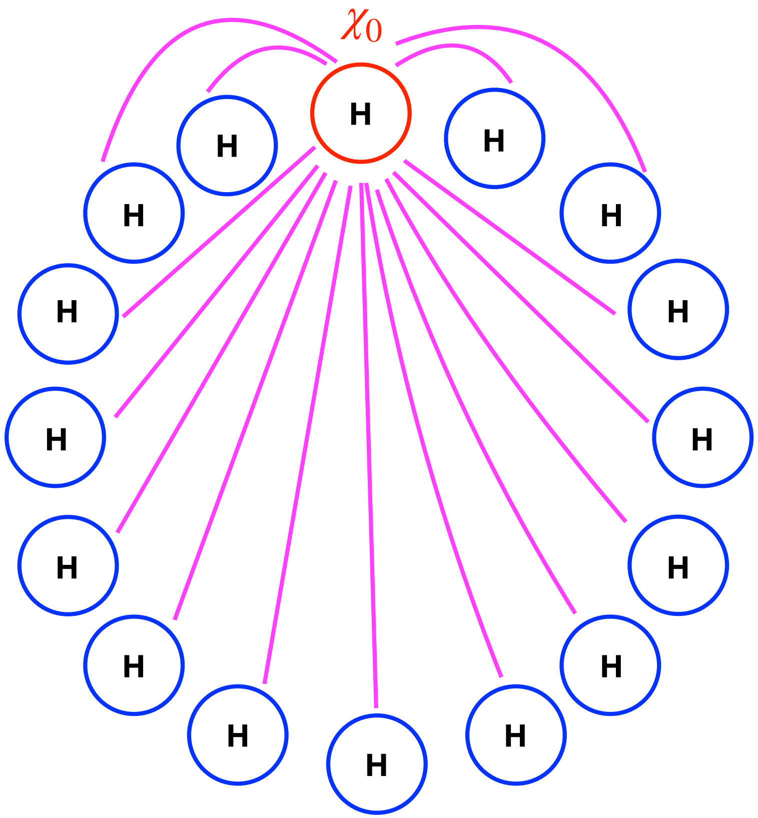

$$\hat{H} = \sum_{IJ} \bar{h}_{IJ}\, \hat{c}_I^\dagger \hat{c}_J$$

Localised representation

Energy contributions involving the impurity

$$2\sum_{J} \bar{h}_{0J} \langle \Psi_0 | \hat{c}_0^\dagger \hat{c}_J | \Psi_0 \rangle$$

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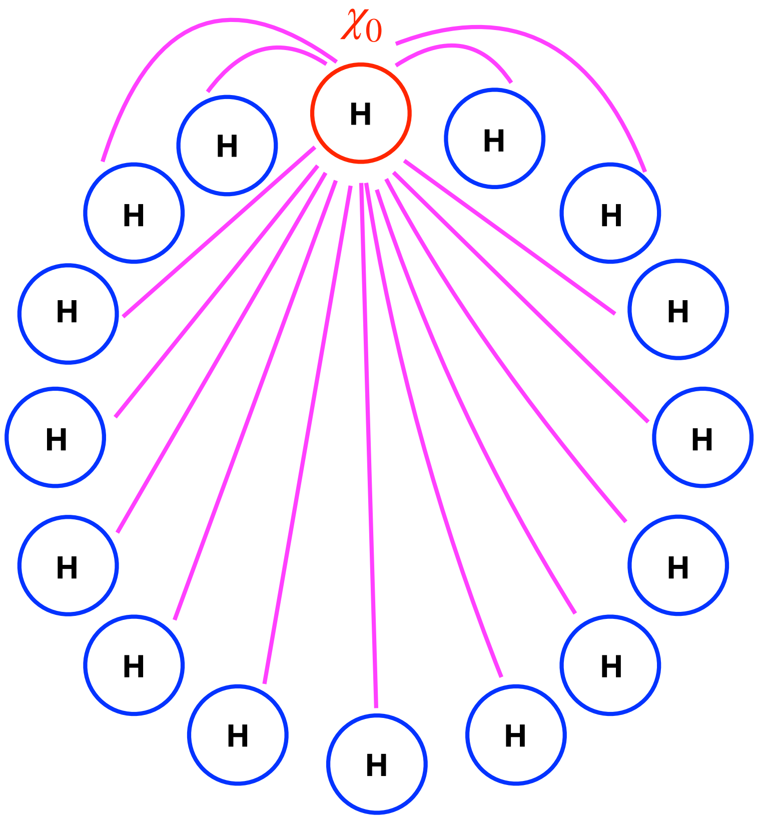

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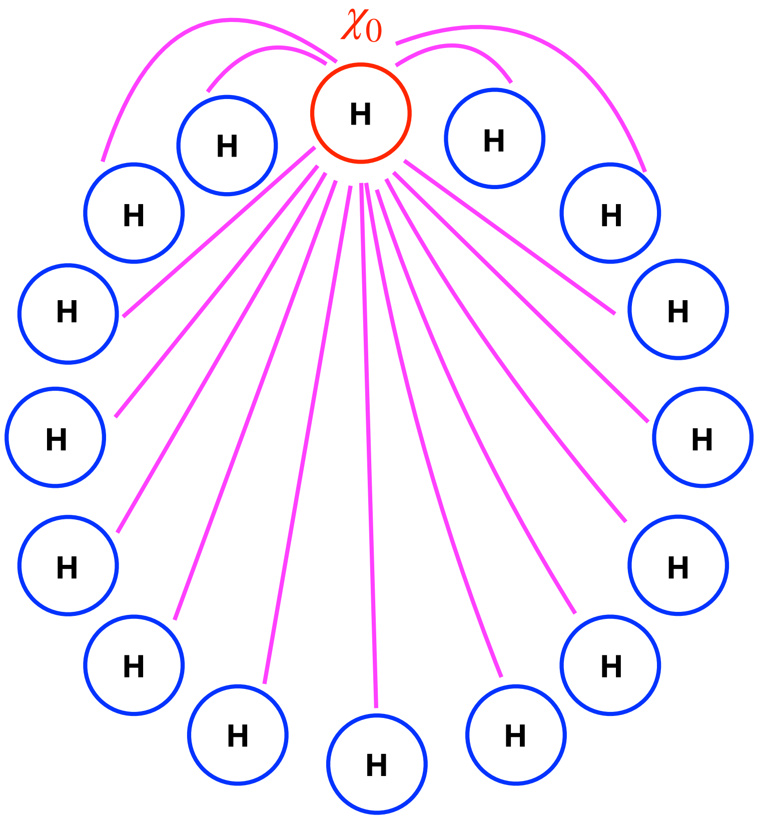
Householder representation

$$= 2 \sum_{K} \underbrace{\left(\sum_{J} \overline{h}_{0J} P_{JK} \right)}_{\tilde{h}_{0K}} \langle \Psi_0 | \hat{d}_0^{\dagger} \hat{d}_K | \Psi_0 \rangle$$

Energy evaluation by fragmentation



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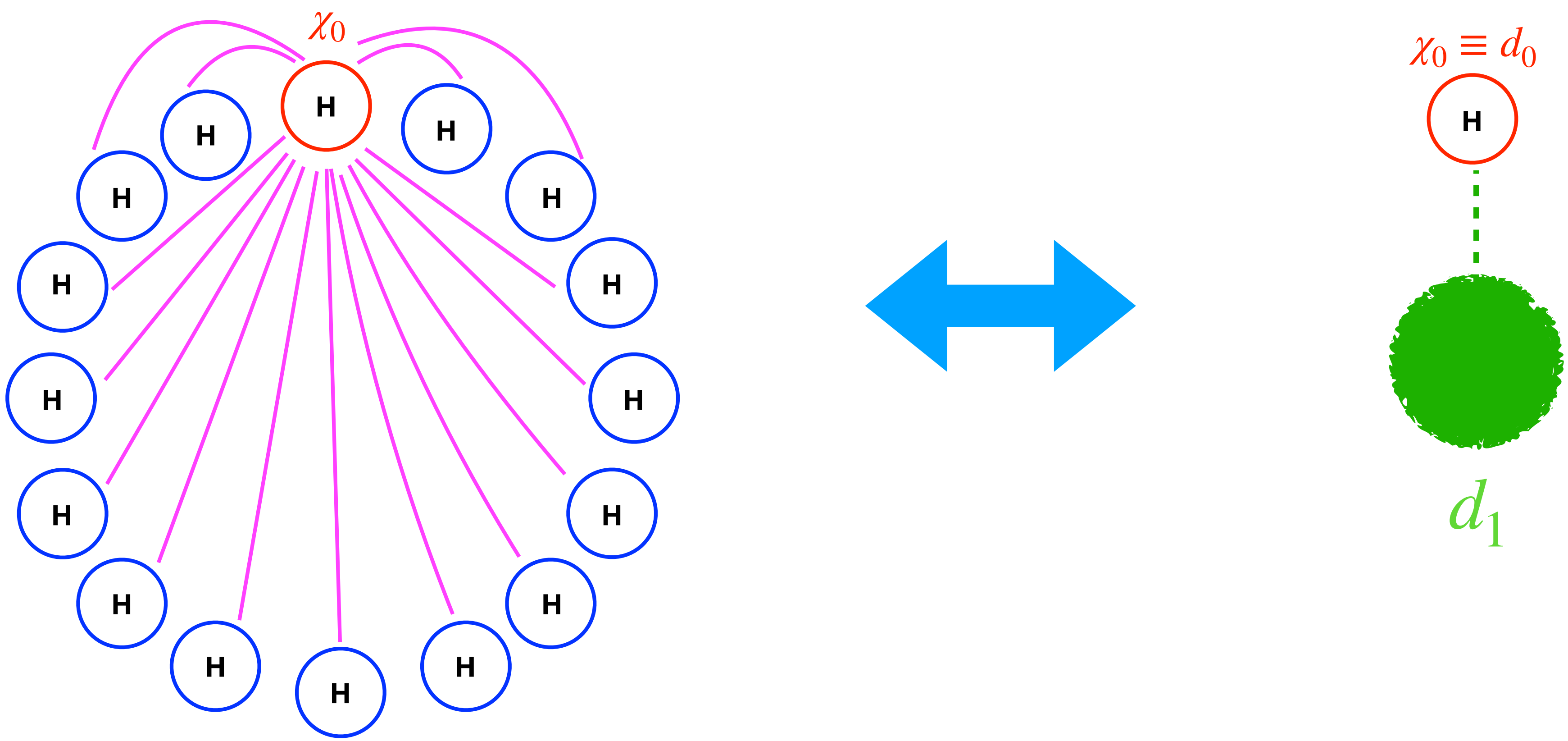
$$= 2 \sum_K \left(\sum_J \bar{h}_{0J} P_{JK} \right) \langle \Psi_0 | \hat{d}_0^\dagger \hat{d}_K | \Psi_0 \rangle$$

$$= 2 \sum_K \tilde{h}_{0K} \tilde{\gamma}_{0K} = 2 \left(\tilde{h}_{00} \tilde{\gamma}_{00} + \tilde{h}_{01} \tilde{\gamma}_{01} \right)$$

Energy evaluation by fragmentation



Determined from the cluster

$$2\sum_J \bar{h}_{0J}\langle\Psi_0|\hat{c}_0^\dagger\hat{c}_J|\Psi_0\rangle = 2\left(\tilde{h}_{00}\,\tilde{\gamma}_{00} + \tilde{h}_{01}\,\tilde{\gamma}_{01}\right)$$

Approximate embedding for interacting electrons

The present embedding approach is useless for non-interacting electrons (!)

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We need

$$\gamma_{IJ} = \gamma_{IJ}^{loc} = \sum_{P}^{occupied\ spin-MOs} C_{IP} C_{JP}$$

Approximate embedding for interacting electrons

We have to solve the Schrödinger equation for the ***full system****!*

We need $$\gamma_{IJ} = \gamma_{IJ}^{loc} = \sum_{P}^{occupied\ spin-MOs} C_{IP} C_{JP}$$

Approximate embedding for interacting electrons

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Nevertheless, the Householder orbitals that have been constructed for non-interacting electrons can be reused as is for performing an (approximate) interacting embedding.

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$$\hat{H} \equiv \sum_{PQ} \tilde{h}_{PQ} \hat{d}^{\dagger}_{P} \hat{d}_{Q} + \frac{1}{2} \sum_{PQRS} \tilde{g}_{PQRS} \hat{d}^{\dagger}_{P} \hat{d}^{\dagger}_{Q} \hat{d}_{S} \hat{d}_{R}$$

*Full Hamiltonian in the **Householder spin-orbitals representation***

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Full Hamiltonian in the Householder spin-orbitals representation

*Determined from the **non-interacting** Hamiltonian*

Approximate embedding for interacting electrons

Nevertheless, the Householder orbitals that have been constructed for non-interacting electrons can be reused as is for performing an (approximate) interacting embedding.

In this case, electron repulsions are taken into account within the Householder cluster.

$$\hat{H} \equiv \sum_{PQ} \tilde{h}_{PQ} \hat{d}^{\dagger}_{P} \hat{d}_{Q} + \frac{1}{2} \sum_{PQRS} \tilde{g}_{PQRS} \hat{d}^{\dagger}_{P} \hat{d}^{\dagger}_{Q} \hat{d}_{S} \hat{d}_{R}$$

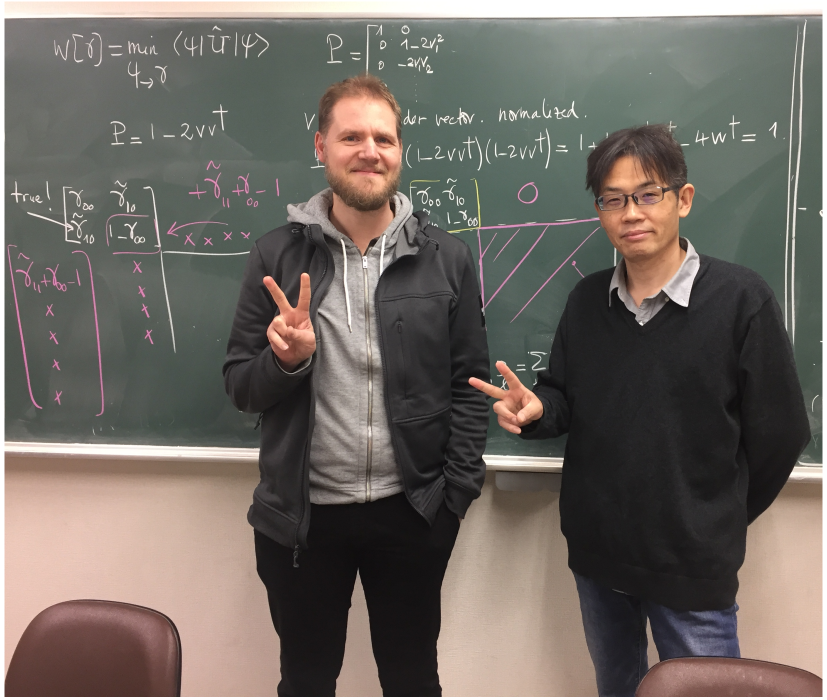
Projection onto the cluster

$$\hat{H}^{\mathscr{C}} \equiv \sum_{P,Q \in \mathscr{C}} \tilde{h}_{PQ} \hat{d}^{\dagger}_{P} \hat{d}_{Q} + \frac{1}{2} \sum_{P,Q,R,S \in \mathscr{C}} \tilde{g}_{PQRS} \hat{d}^{\dagger}_{P} \hat{d}^{\dagger}_{Q} \hat{d}_{S} \hat{d}_{R}$$

Application to the 1D Hubbard model

The “Householder embedding” project

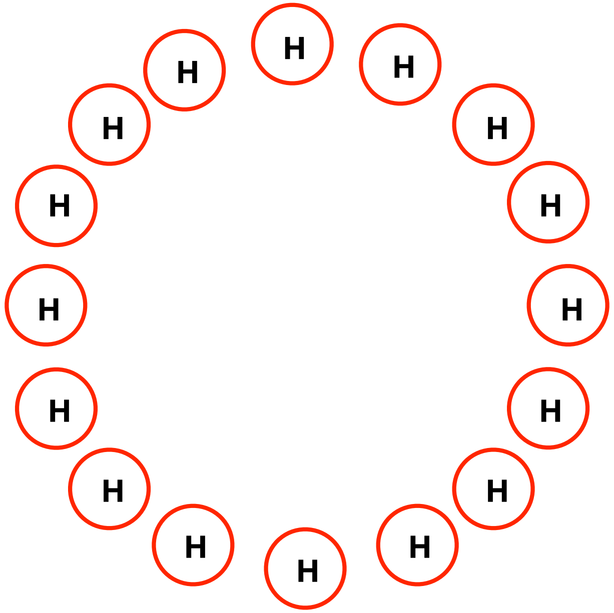
From left to right: ***S. Sekaran*** *(Strasbourg, France),*
M. Saubanère *(Montpellier, France),*
L. Mazouin *(Strasbourg, France), and E.F.*



E.F. and ***M. Tsuchiizu*** *(Nara, Japan).*

A. S. Householder, J. ACM 5, 339 (1958).
S. Sekaran, M. Tsuchiizu, M. Saubanère, and E. Fromager, Phys. Rev. B ***104****, 035121 (2021).*

Prototypical ring of L hydrogen atoms



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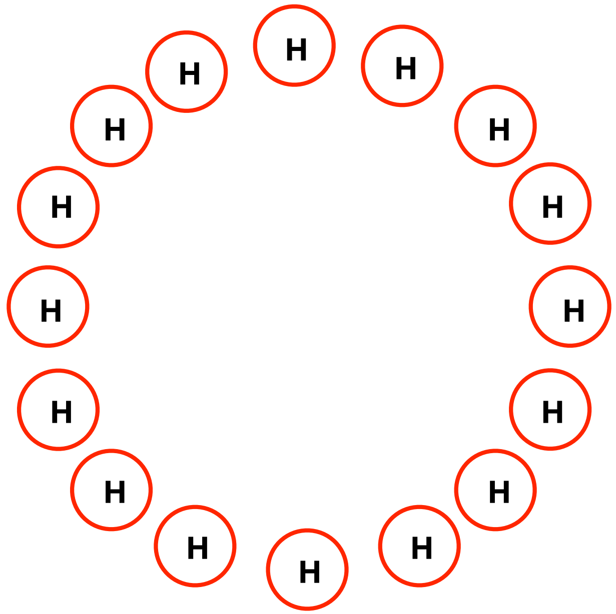

Hubbard model

$$\hat{H} \approx -t \sum_{\sigma=\uparrow,\downarrow} \sum_{i=0}^{L-1} \left(\hat{c}_{i\sigma}^{\dagger} \hat{c}_{(i+1)\sigma} + \hat{c}_{(i+1)\sigma}^{\dagger} \hat{c}_{i\sigma} \right) + U \sum_{i=0}^{L-1} \hat{c}_{i\uparrow}^{\dagger} \hat{c}_{i\downarrow}^{\dagger} \hat{c}_{i\downarrow} \hat{c}_{i\uparrow}$$

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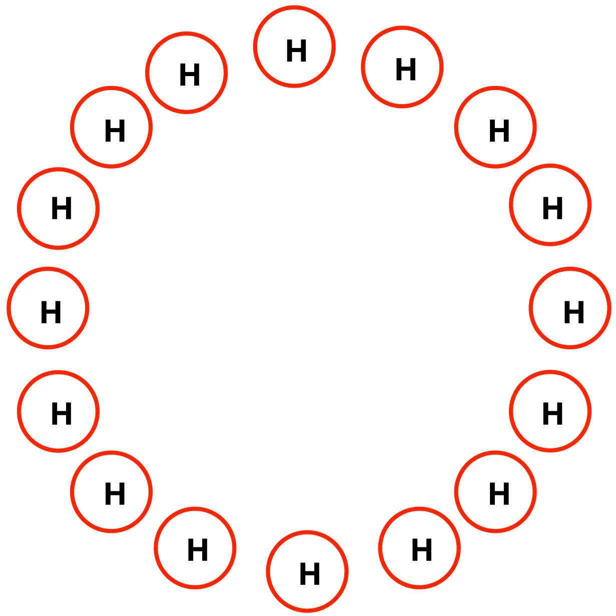

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Hückel model $-t \equiv \beta$

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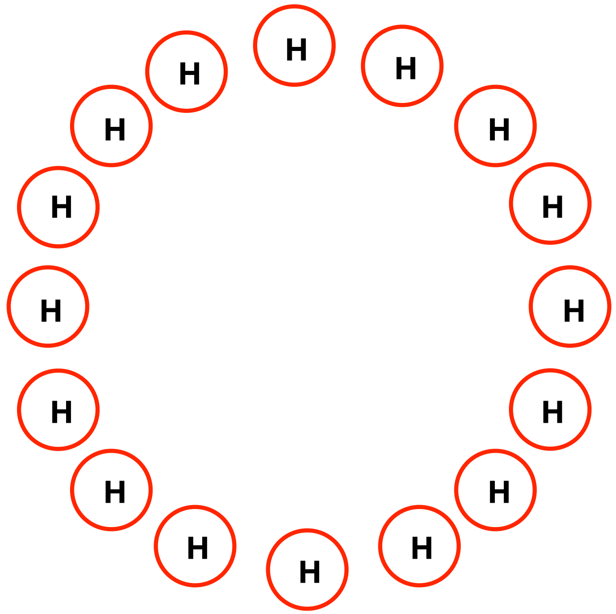

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Two-electron repulsion on each atom only

Prototypical ring of L hydrogen atoms

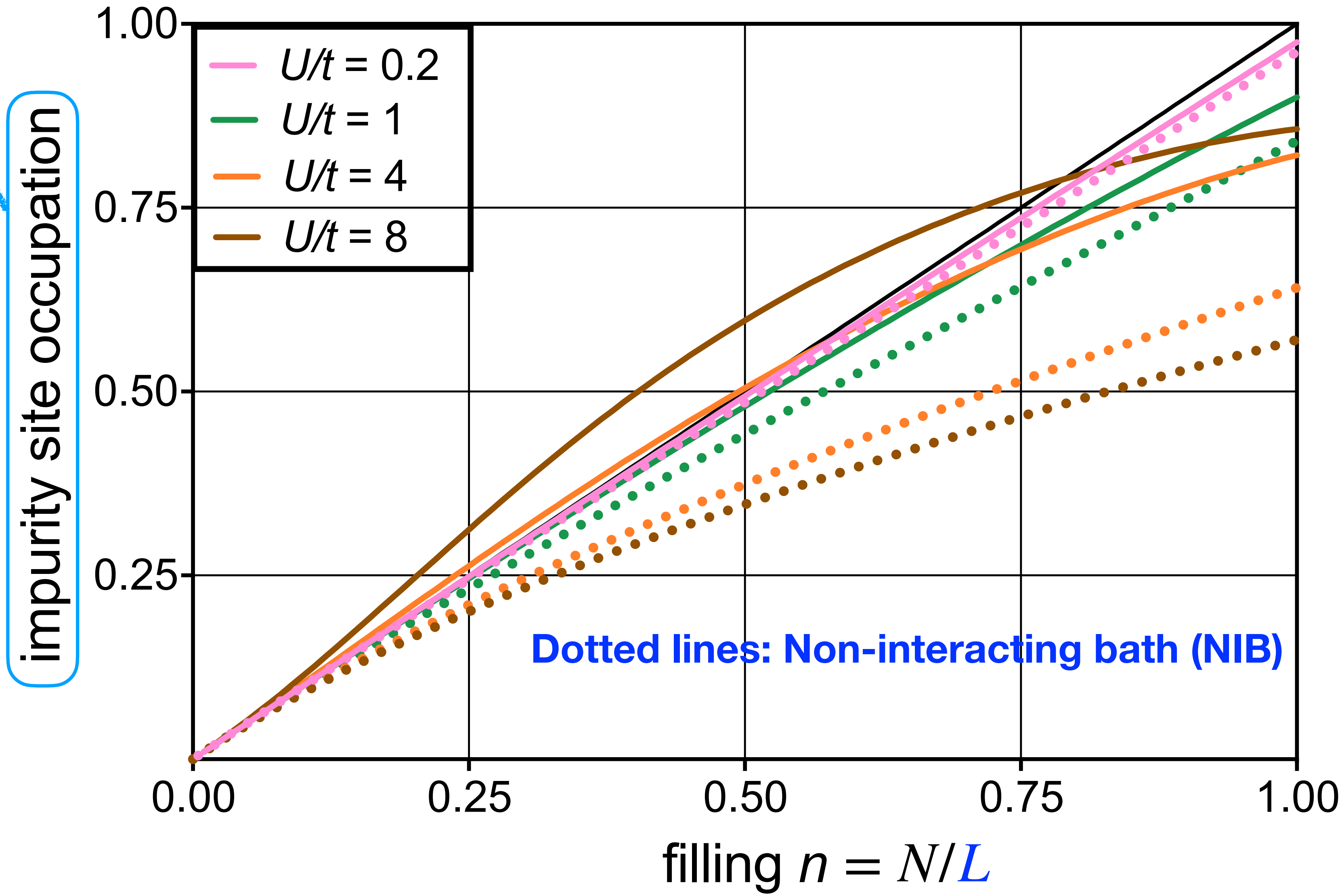
$$U/t \ll 1$$ ***Weakly*** *correlated regime*

$$U/t \gg 1$$ ***Strongly*** *correlated regime*

Hubbard model

$$\hat{H} \approx -t \sum_{\sigma=\uparrow,\downarrow} \sum_{i=0}^{L-1} \left(\hat{c}^\dagger_{i\sigma} \hat{c}_{(i+1)\sigma} + \hat{c}^\dagger_{(i+1)\sigma} \hat{c}_{i\sigma} \right) + U \sum_{i=0}^{L-1} \hat{c}^\dagger_{i\uparrow} \hat{c}^\dagger_{i\downarrow} \hat{c}_{i\downarrow} \hat{c}_{i\uparrow}$$

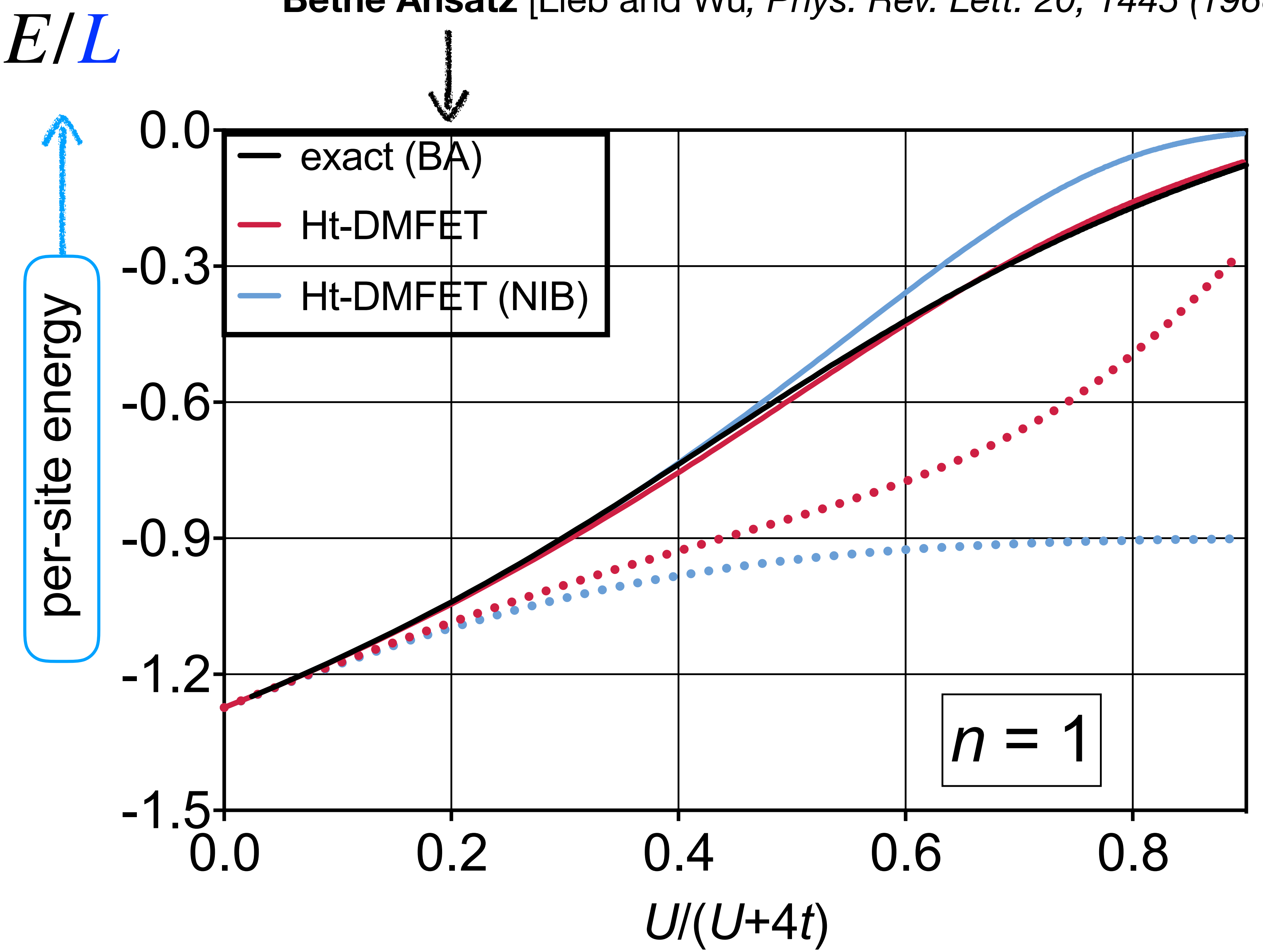
$$\sum_{\sigma=\uparrow,\downarrow} \langle \Psi^{\mathscr{C}} | \hat{c}^{\dagger}_{0\sigma} \hat{c}_{0\sigma} | \Psi^{\mathscr{C}} \rangle$$



$L = 400$ atoms

S. Sekaran, M. Tsuchiizu, M. Saubanère, and E. Fromager, Phys. Rev. B **104**, 035121 (2021).

Bethe Ansatz [Lieb and Wu, *Phys. Rev. Lett. 20, 1445 (1968)*]



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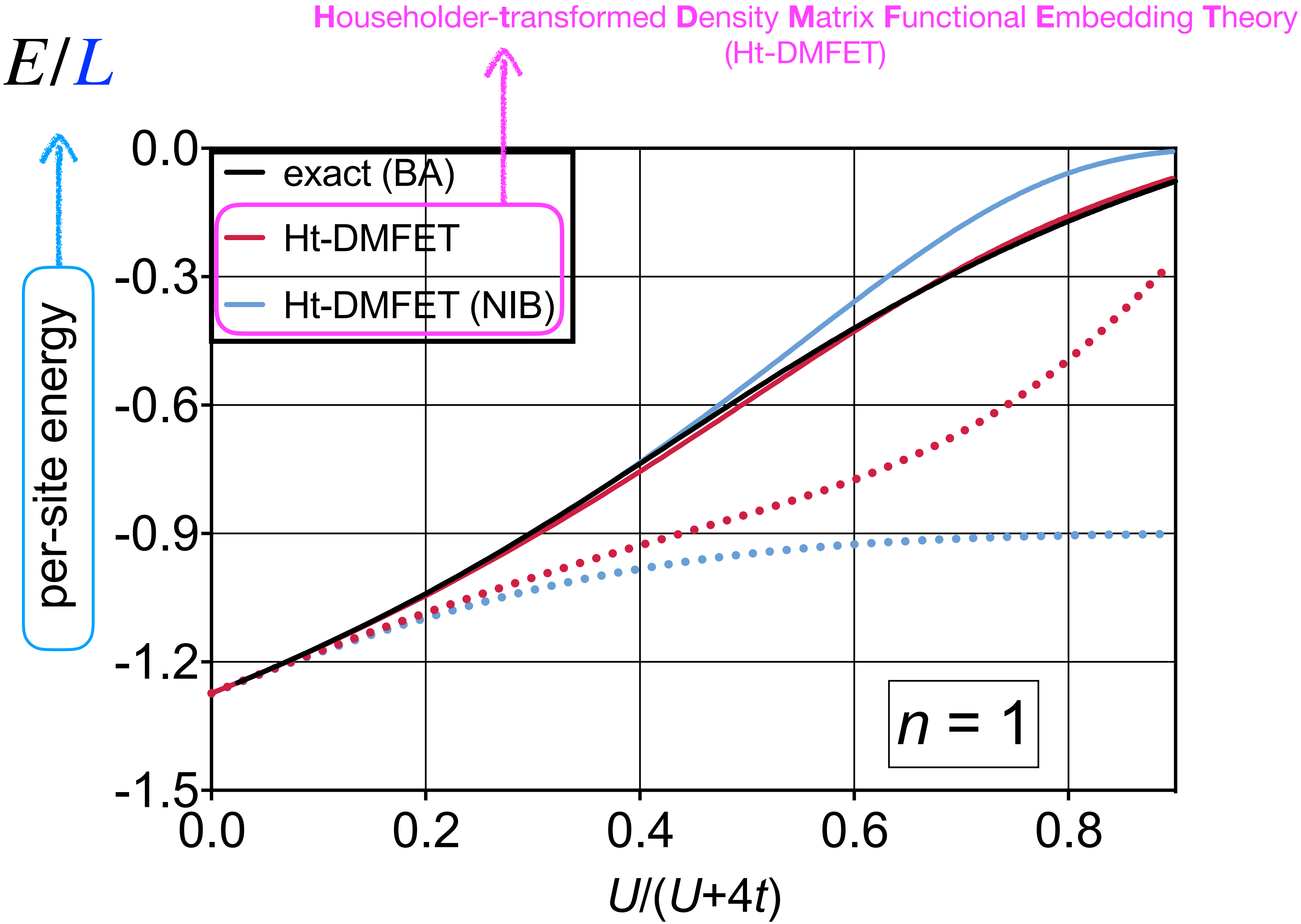

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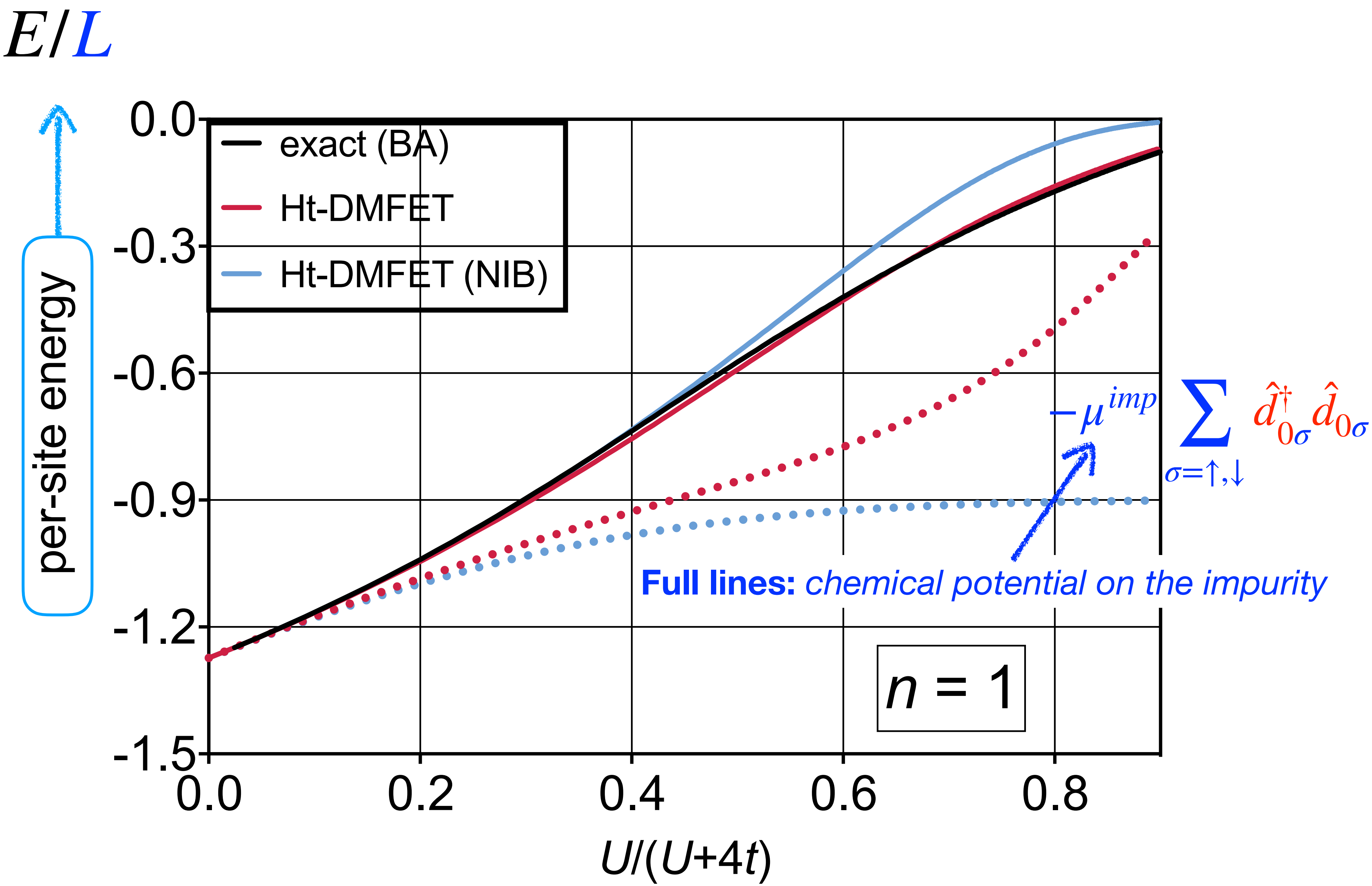

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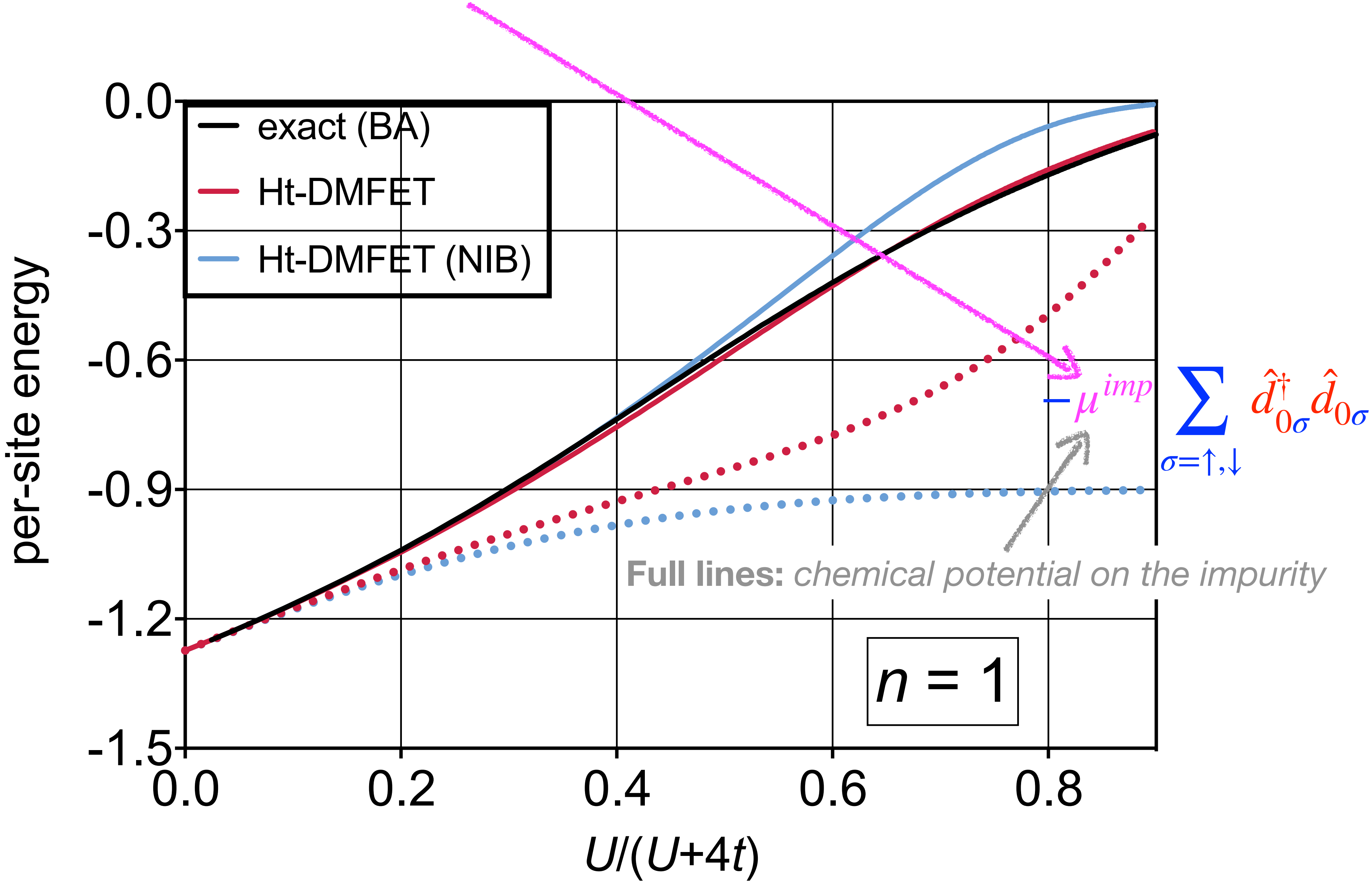
Can be interpreted as an ***approximation to the exact Hxc density-functional potential*** *of the full lattice**



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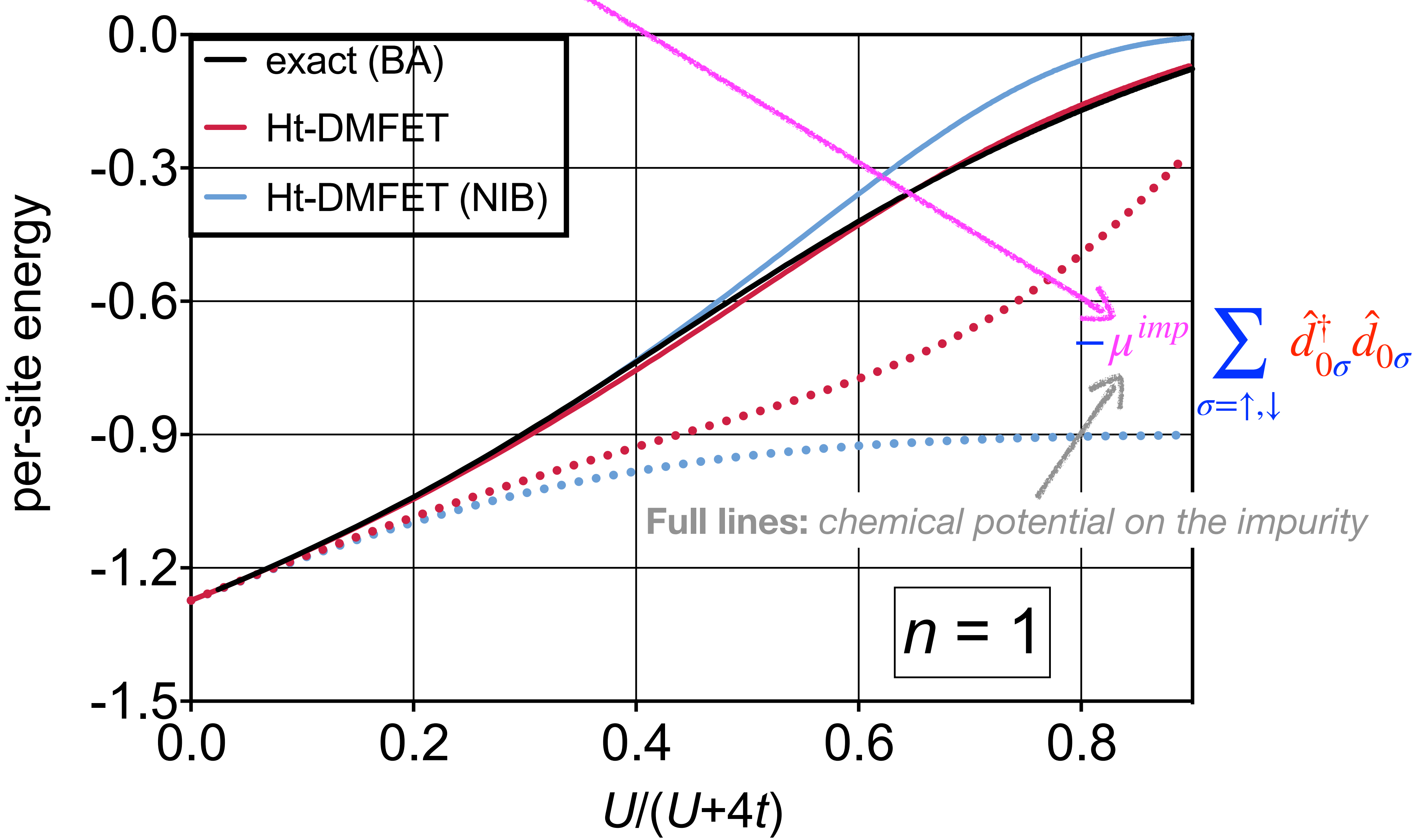
**S. Sekaran, M. Saubanère, and E. Fromager, to be submitted (2022).*

Can be interpreted as an ***approximation to the exact Hxc density-functional potential*** *of the full lattice**

"DFT without density functionals"



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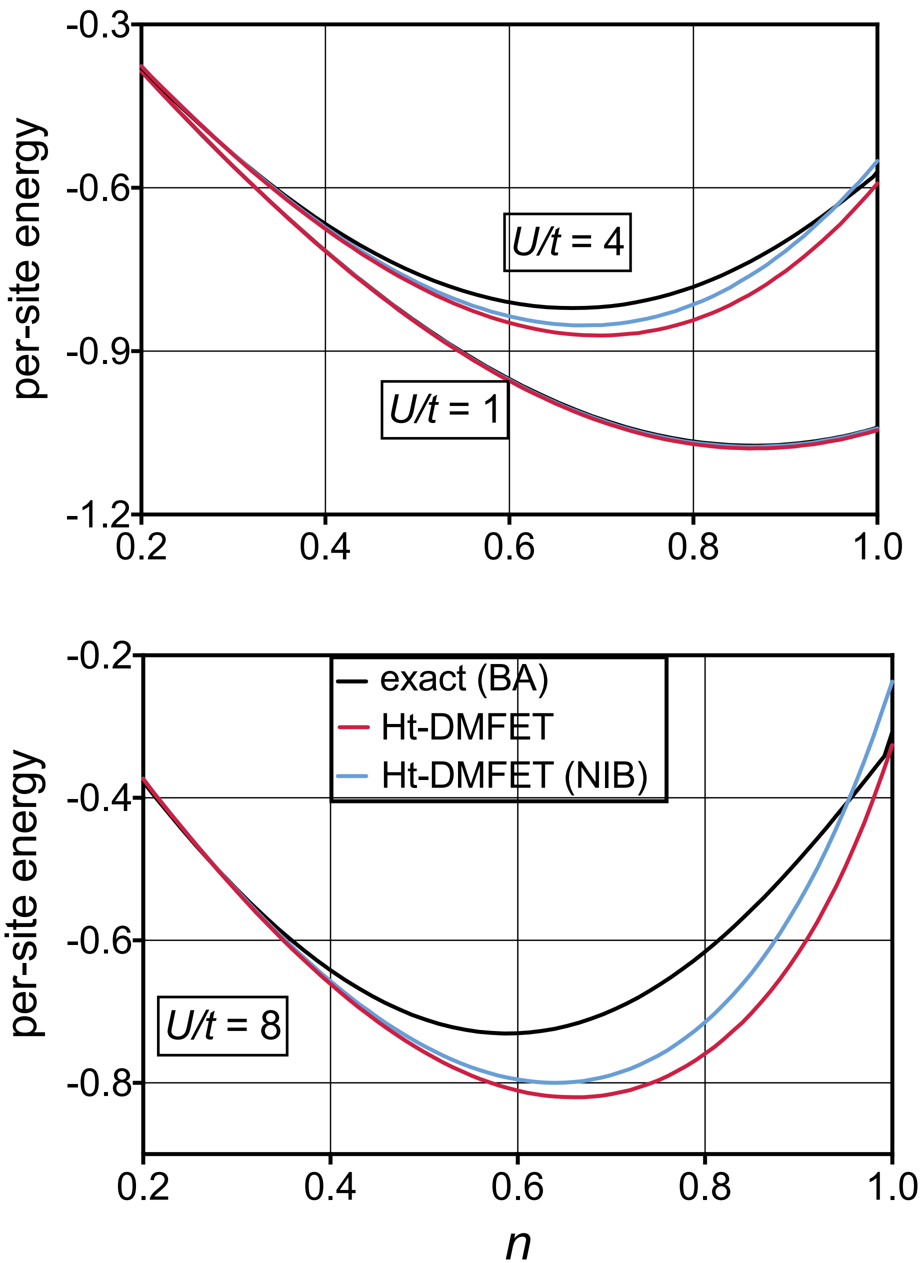
Ht-DMFET per-site energies away from half-filling ($n < 1$)



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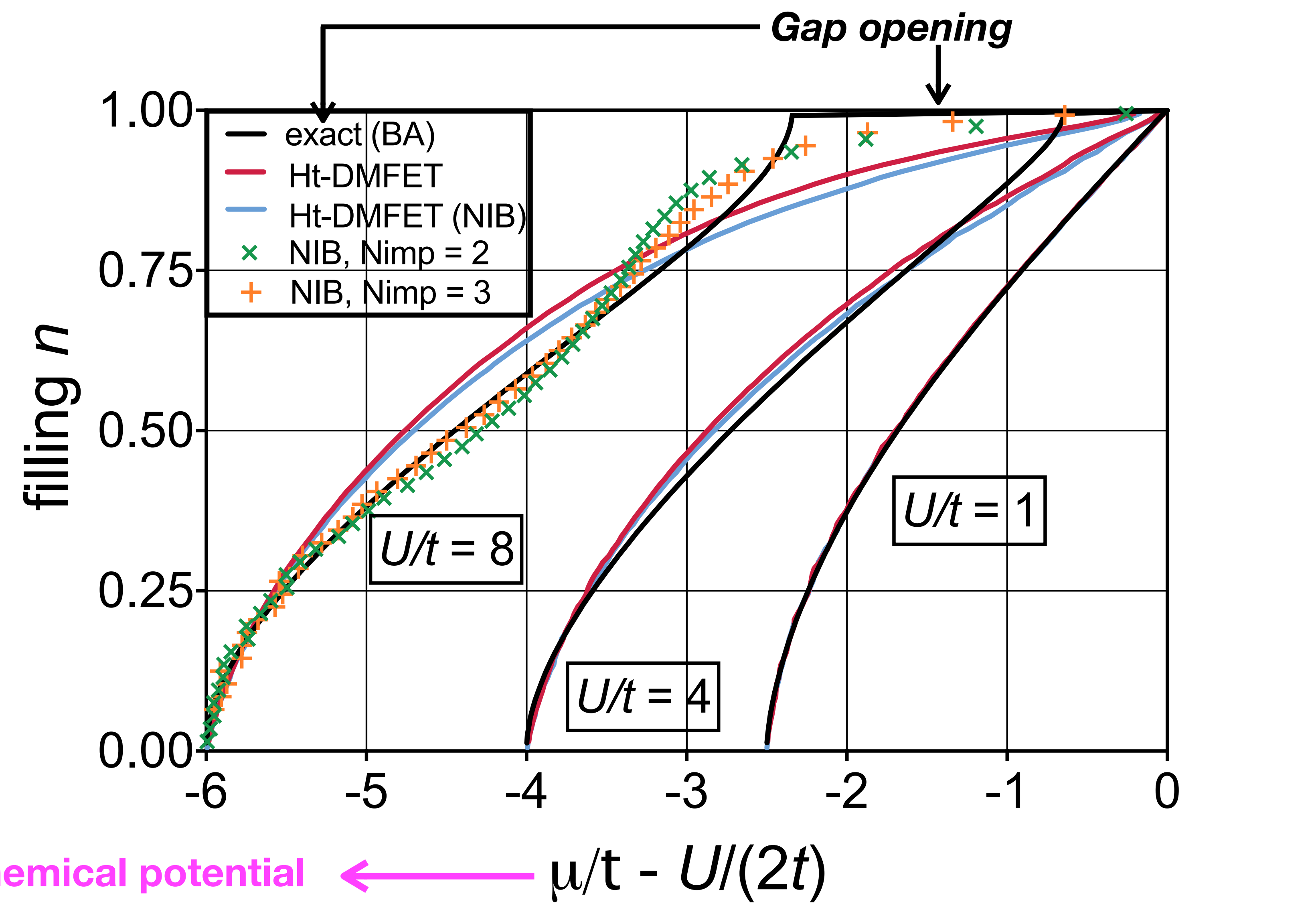
Mott-Hubbard density-driven transition and multiple impurities



Chemical potential ←

$$\mu \equiv \mu(n) = \frac{1}{L}\frac{\partial E(n)}{\partial n}$$

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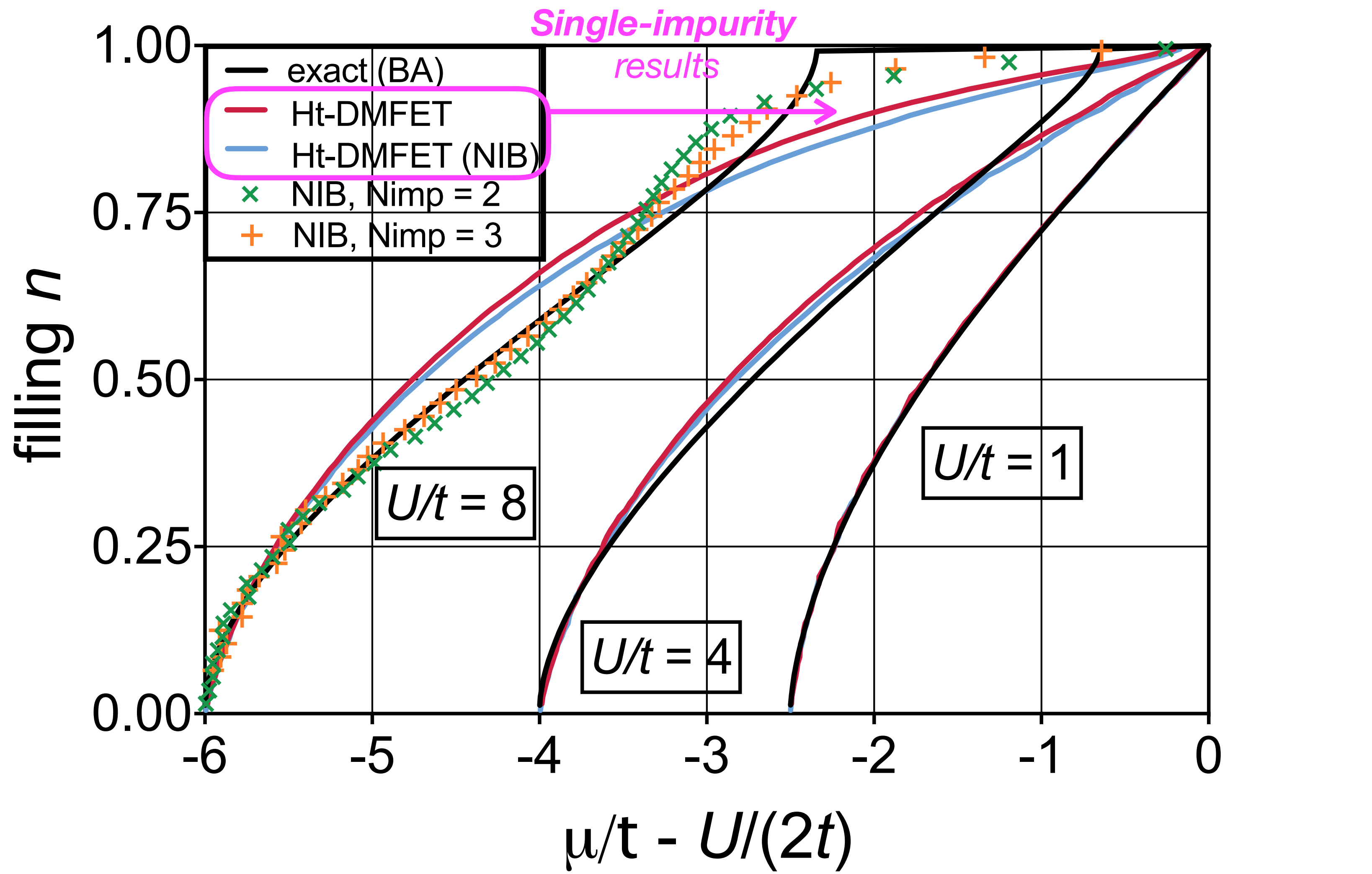

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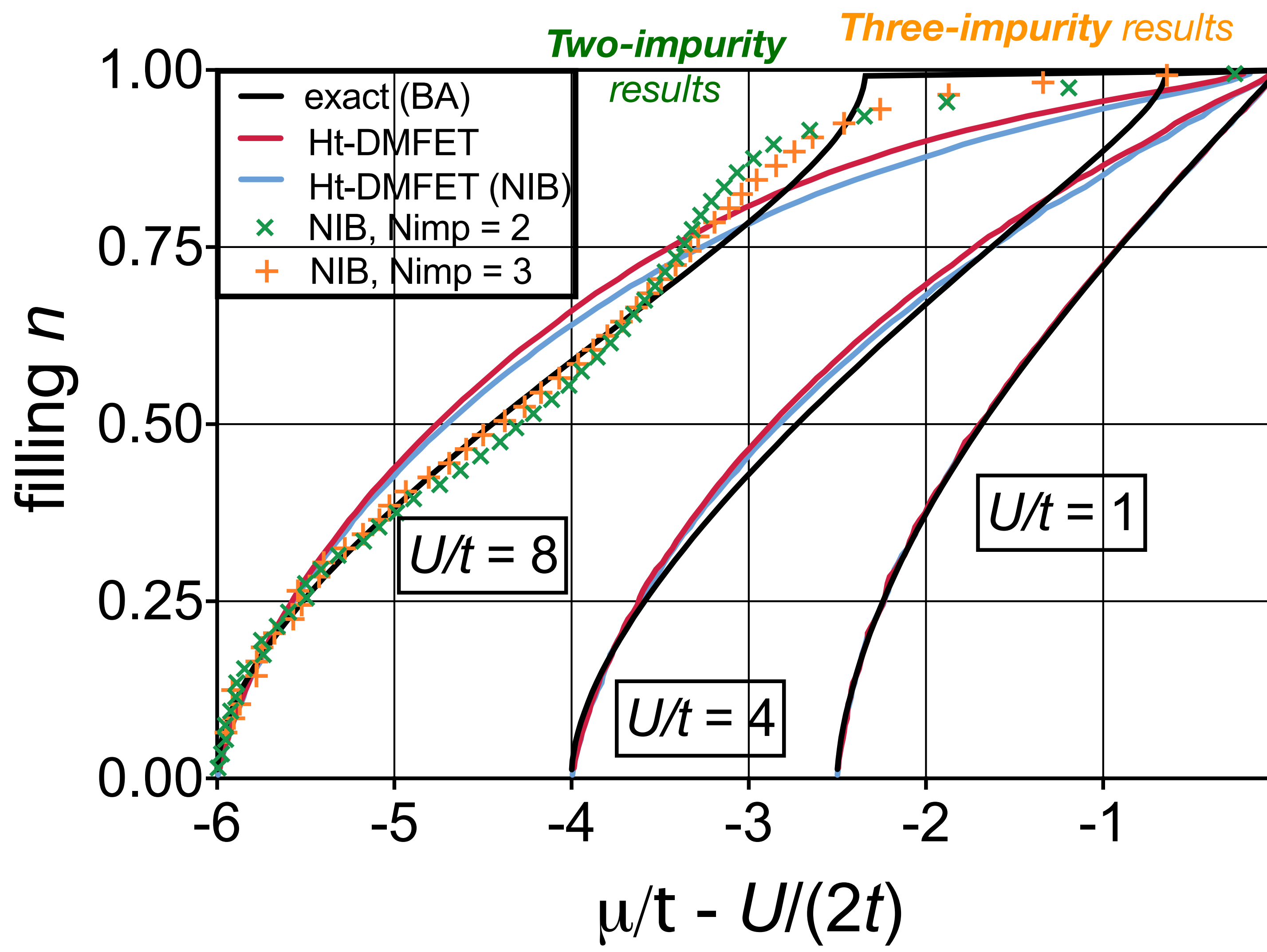

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