Householder transformed density matrix functional theory

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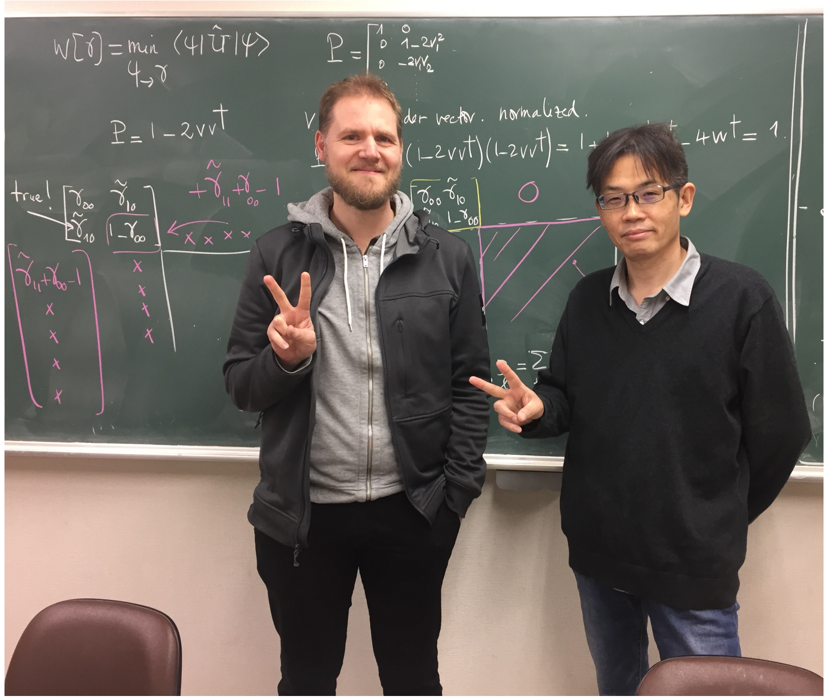
[b]*Department of Physics, Nara Women's University, Nara, Japan*
[c]*Institut Charles Gerhardt, CNRS/Université de Montpellier, France*

"Tensor product methods for strongly correlated molecular systems" virtual focus workshop
MPIPKS, Dresden, Germany

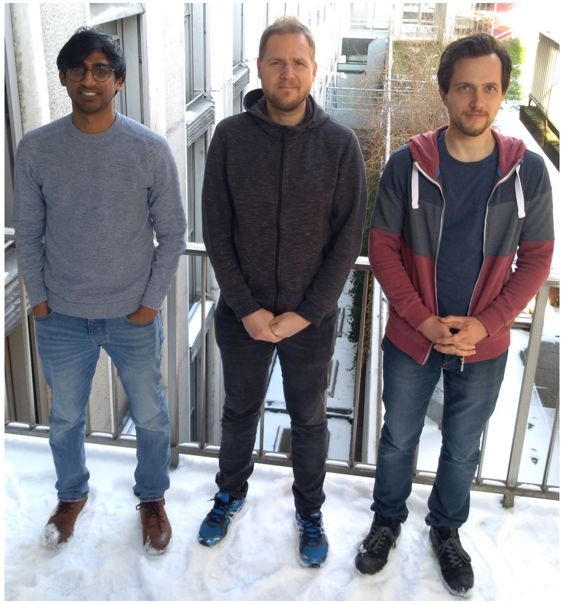
10/03/2021

The Householder team

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).*

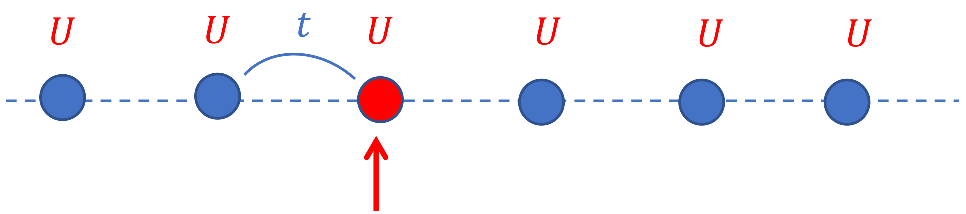


On the right: ***Filip Cernatic*** *(PhD student, Strasbourg).*

Density matrix approach to quantum embedding

Condensed Matter Physics | **Quantum Chemistry**

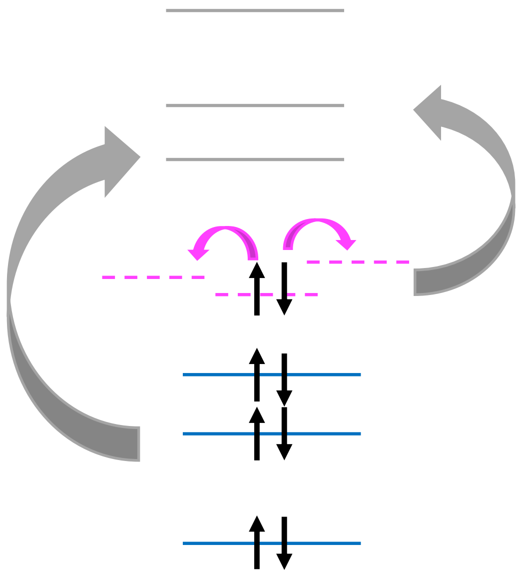
Hubbard model



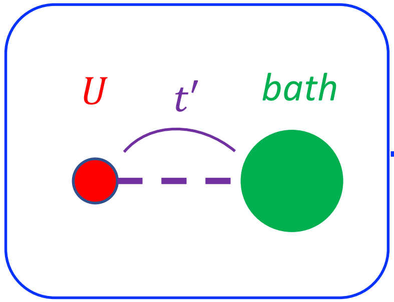
To-be-embedded atomic site (impurity)

$$\hat{H}\,\Psi = E\,\Psi$$

Full system

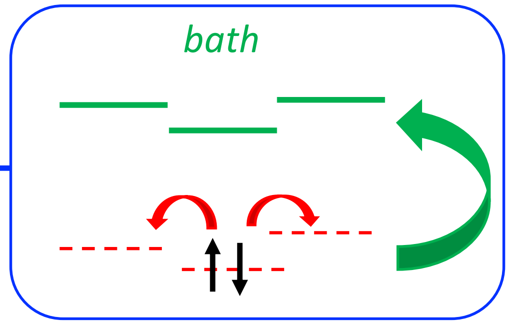


The "density matrix embedding theory" (DMET) picture

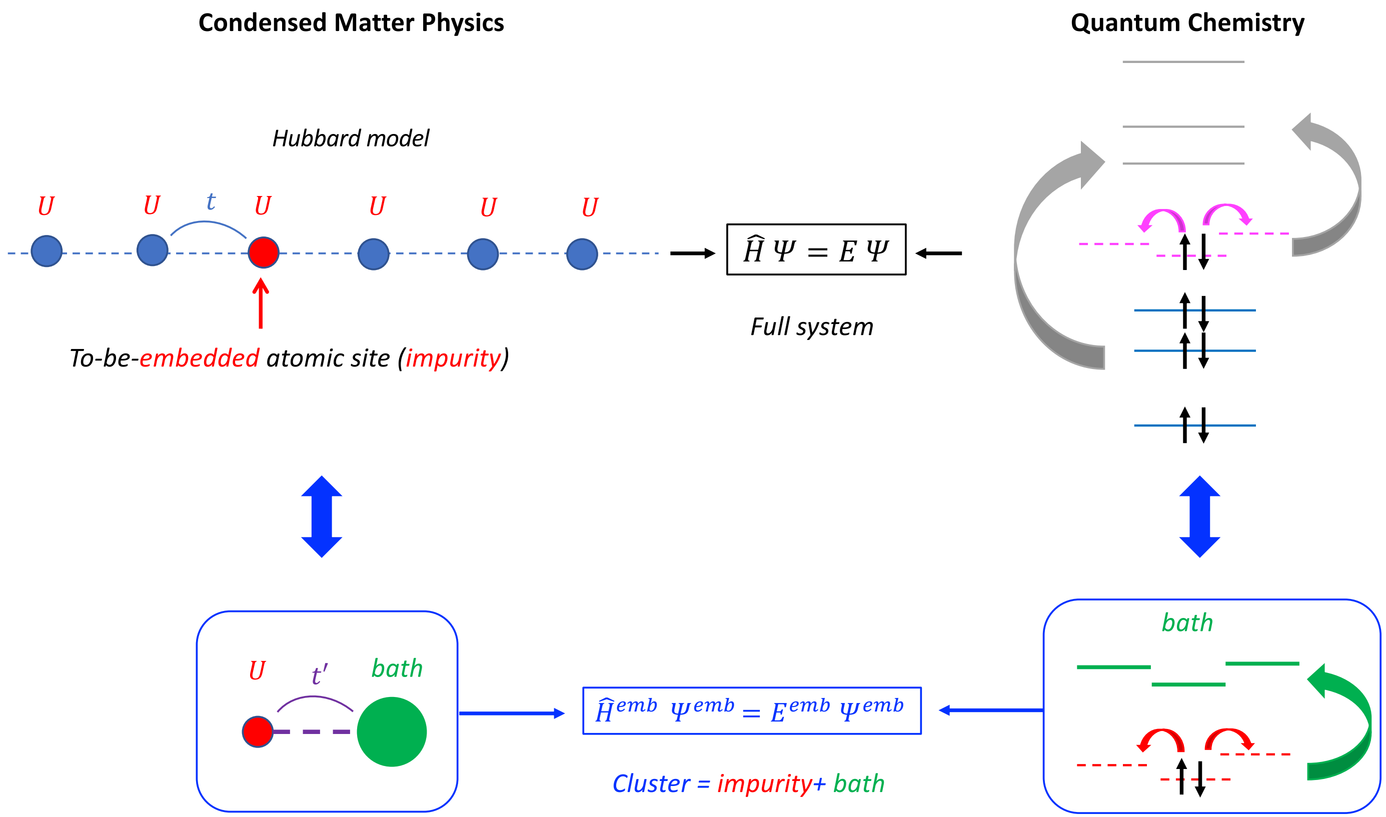


$$\hat{H}^{emb}\ \Psi^{emb} = E^{emb}\ \Psi^{emb}$$

Cluster = impurity+ bath



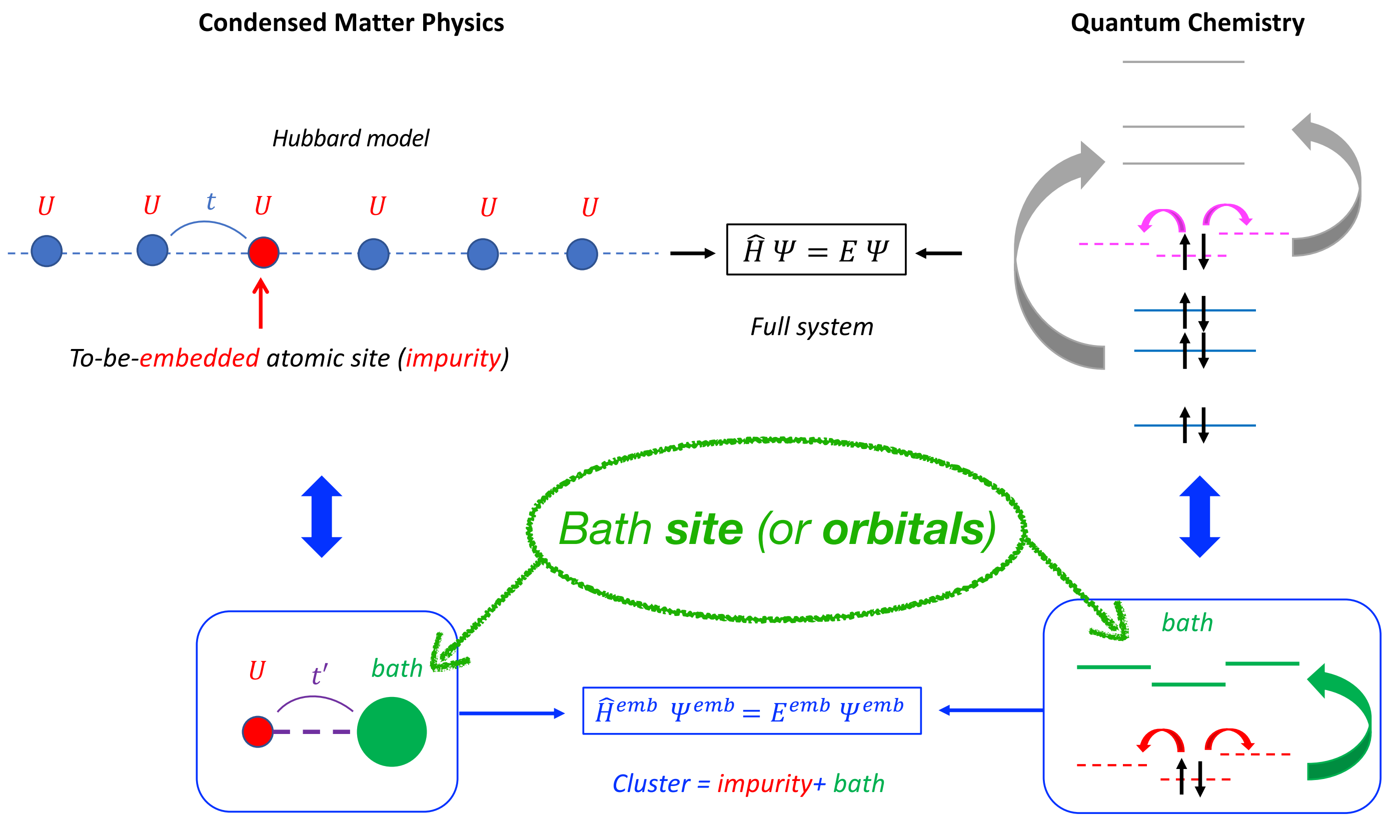
G. Knizia and G. K.-L. Chan, Phys. Rev. Lett. **109**, 186404 (2012).



Local evaluation of reduced density matrices, ***from the cluster:***

$$\langle \hat{c}^\dagger_{i\sigma}\hat{c}_{j\sigma}\rangle_{\Psi} \approx \langle \hat{c}^\dagger_{i\sigma}\hat{c}_{j\sigma}\rangle_{\Psi^{emb}}$$

$$\langle \hat{c}^\dagger_{i\sigma}\hat{c}^\dagger_{j\tau}\hat{c}_{l\tau}\hat{c}_{k\sigma}\rangle_{\Psi} \approx \langle \hat{c}^\dagger_{i\sigma}\hat{c}^\dagger_{j\tau}\hat{c}_{l\tau}\hat{c}_{k\sigma}\rangle_{\Psi^{emb}}$$



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Approximate local properties

(per-site energy, double occupation)

Systematically improvable embedding within a single-particle bath picture

PHYSICAL REVIEW B **103**, 085131 (2021)

Fully algebraic and self-consistent effective dynamics in a static quantum embedding

P. V. Sriluckshmy, Max Nusspickel, Edoardo Fertitta, and George H. Booth *

Department of Physics, King's College London, Strand, London WC2R 2LS, United Kingdom

(Received 10 December 2020; revised 3 February 2021; accepted 5 February 2021; published 22 February 2021)

Quantum embedding approaches involve the self-consistent optimization of a local fragment of a strongly correlated system, entangled with the wider environment. The 'energy-weighted' density matrix embedding theory (EwDMET) was established recently as a way to systematically control the resolution of the fragment-environment coupling and allow for true quantum fluctuations over this boundary to be self-consistently optimized within a fully static framework. In this work, we reformulate the algorithm to ensure that EwDMET can be considered equivalent to an optimal and rigorous truncation of the self-consistent dynamics of dynamical mean-field theory (DMFT). A practical limitation of these quantum embedding approaches is often a numerical fitting of a self-consistent object defining the quantum effects. However, we show here that in this formulation, all numerical fitting steps can be entirely circumvented, via an effective Dyson equation in the space of truncated dynamics. This provides a robust and analytic self-consistency for the method, and an ability to systematically and rigorously converge to DMFT from a static, wave function perspective. We demonstrate that this improved approach can solve the correlated dynamics and phase transitions of the Bethe lattice Hubbard model in infinite dimensions, as well as one- and two-dimensional Hubbard models where we clearly show the benefits of this rapidly convergent basis for correlation-driven fluctuations. This systematically truncated description of the effective dynamics of the problem also allows access to quantities such as Fermi liquid parameters and renormalized dynamics, and demonstrates a numerically efficient, systematic convergence to the zero-temperature dynamical mean-field theory limit.

DOI: 10.1103/PhysRevB.103.085131

Systematically improvable embedding within a single-particle bath picture

- *We want to follow a different path.*
- *We want the embedding to remain formally static, like in the original DMET*[1].
- *Our strategy: Think the embedding as a functional of the (one-electron) density matrix*[2]

$$\gamma_{ij} \equiv \langle \hat{c}^{\dagger}_{i\sigma} \hat{c}_{j\sigma} \rangle$$

- *Correlation may be introduced into the bath through the density matrix.*

[1] *G. Knizia and G. K.-L. Chan, Phys. Rev. Lett.* ***109****, 186404 (2012).*
[2] *S. Sekaran, M. Tsuchiizu, M. Saubanère, and E. Fromager,* ***arXiv:2103.04194*** *(2021).*

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*In the following, I will discuss the embedding a **single impurity** in a **1D Hubbard lattice.***

[1] *G. Knizia and G. K.-L. Chan, Phys. Rev. Lett.* ***109****, 186404 (2012).*
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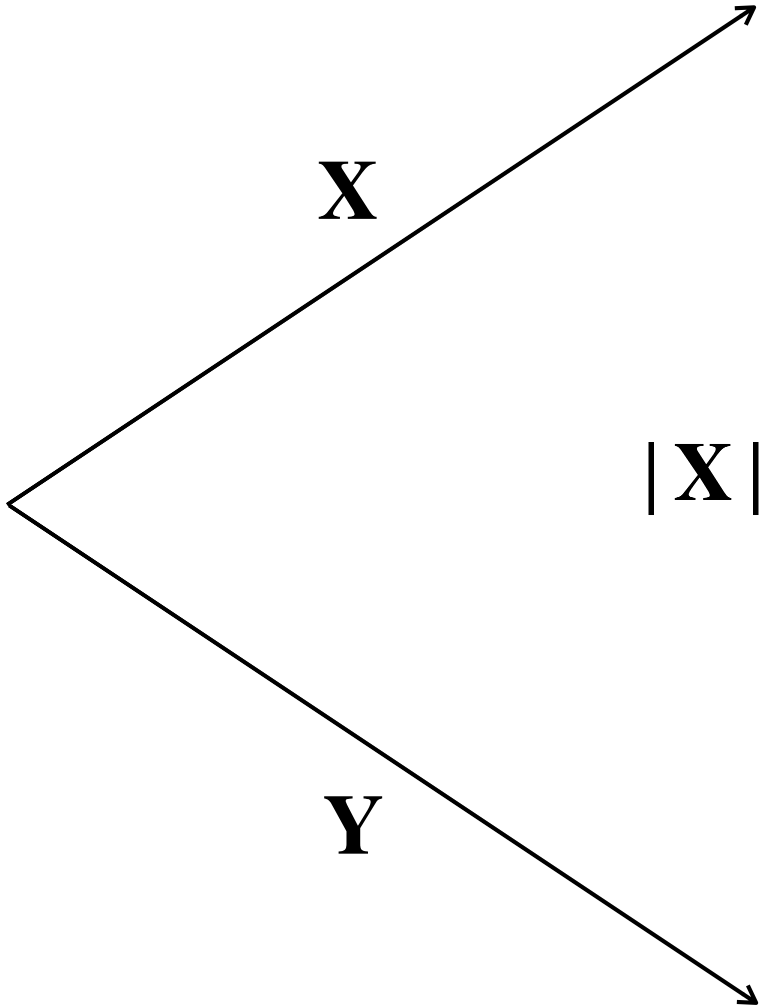
The Householder transformation



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

A. S. Householder, J. ACM 5, 339 (1958).
S. Sekaran, M. Tsuchiizu, M. Saubanère, and E. Fromager, ***arXiv:2103.04194*** *(2021).*

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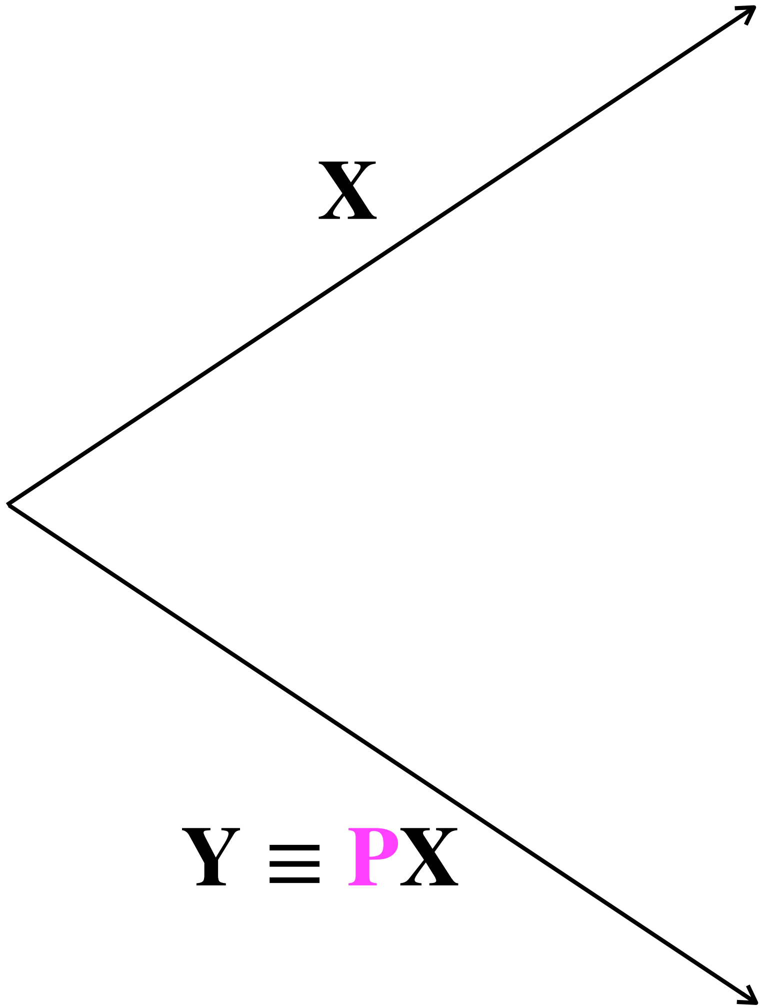

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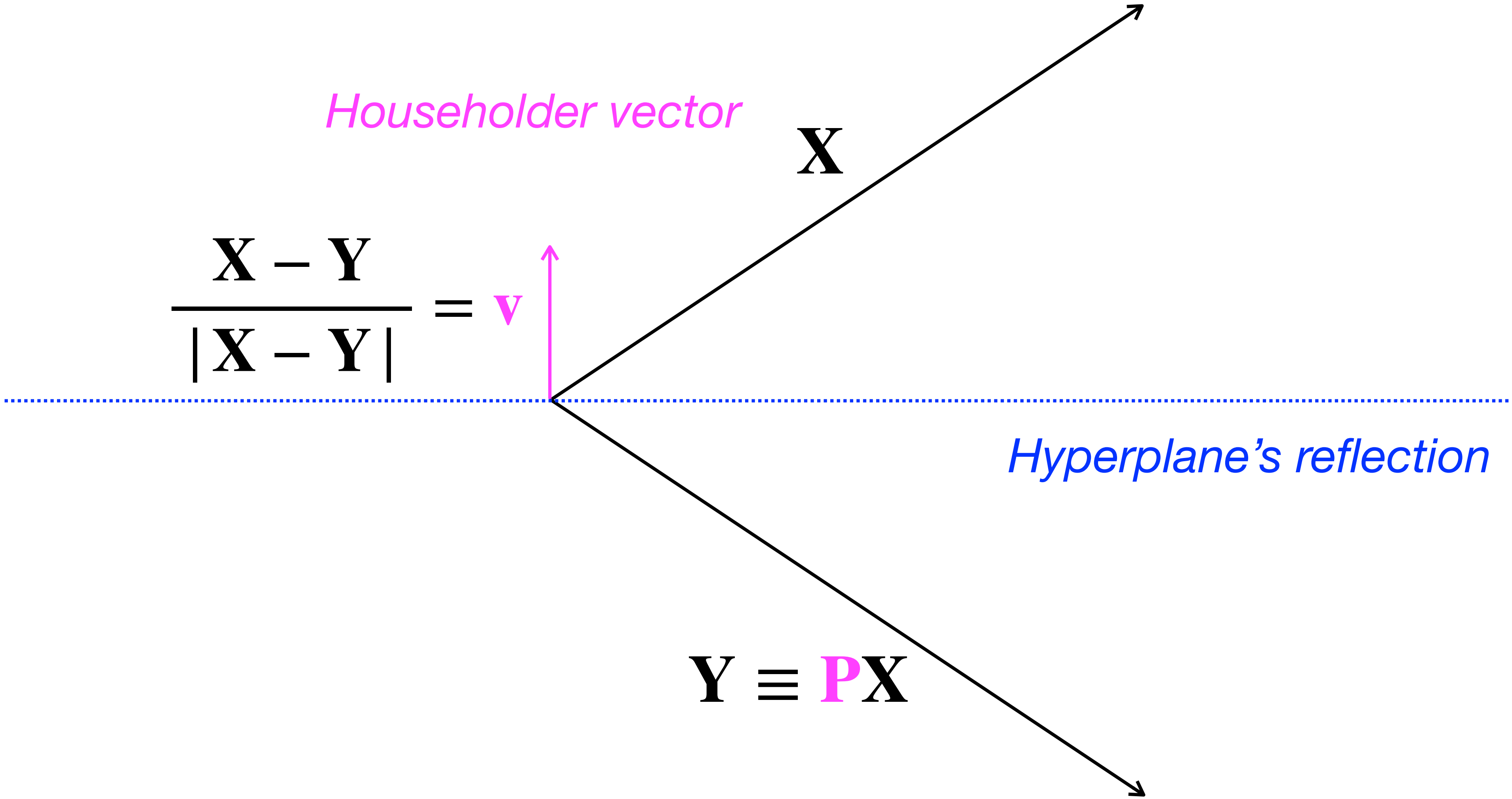

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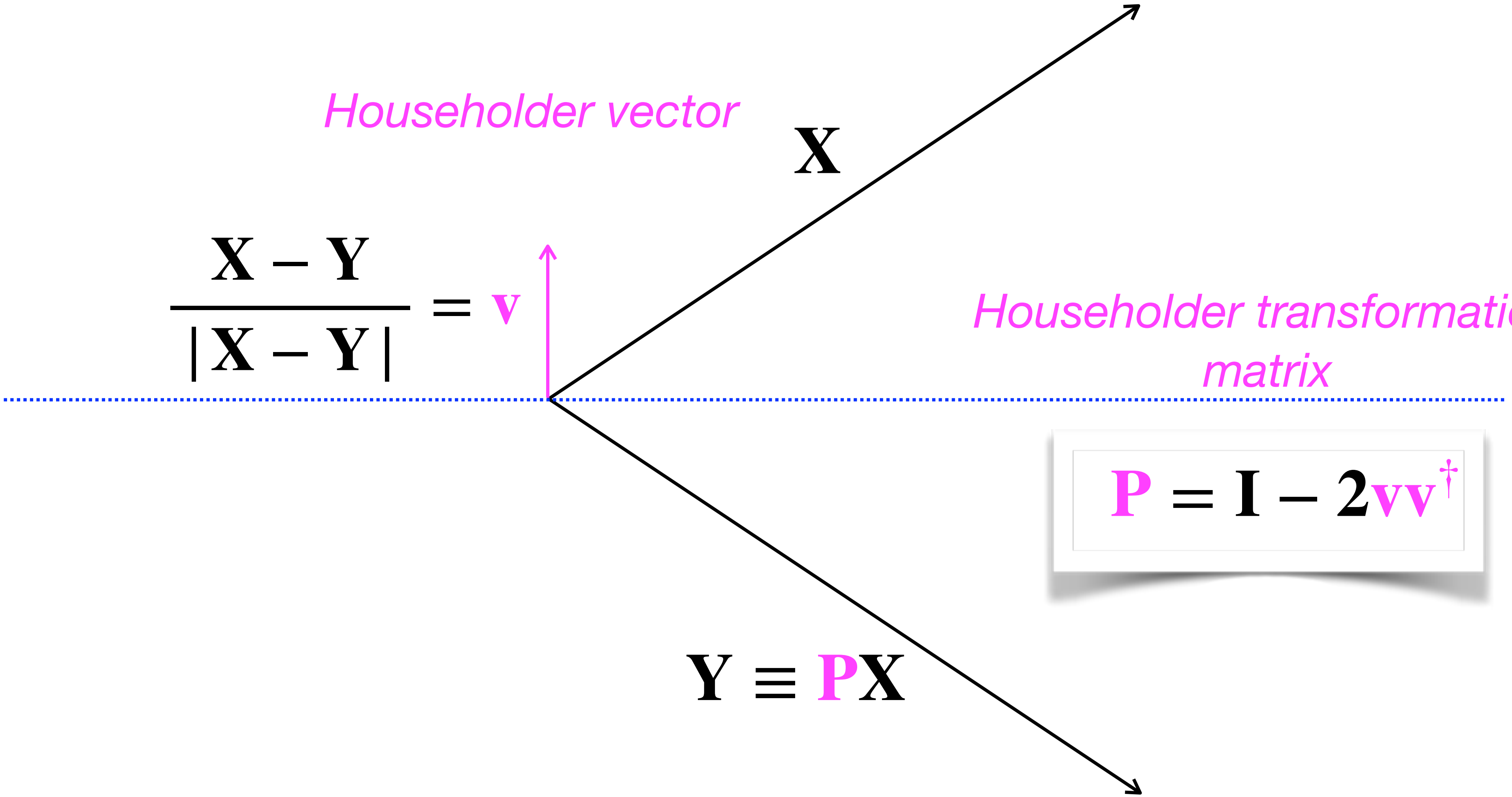


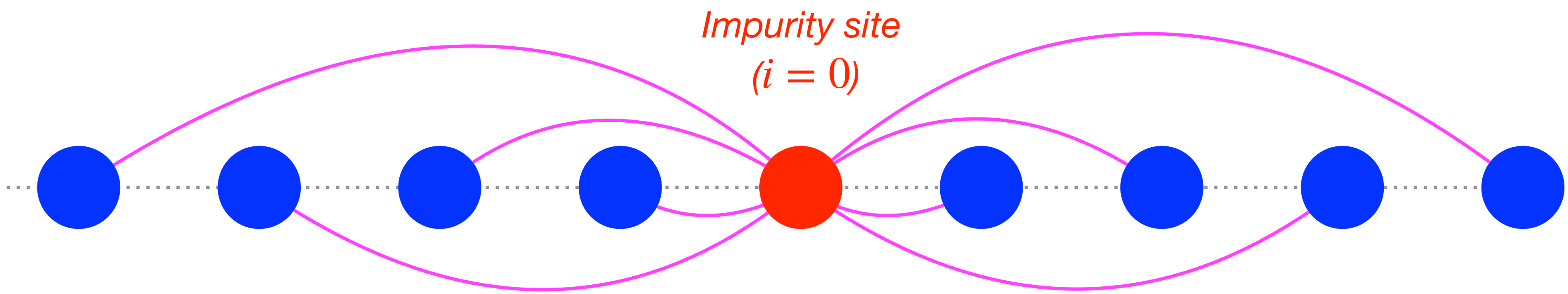
$$\frac{\mathbf{X}-\mathbf{Y}}{|\mathbf{X}-\mathbf{Y}|} = \mathbf{v}$$

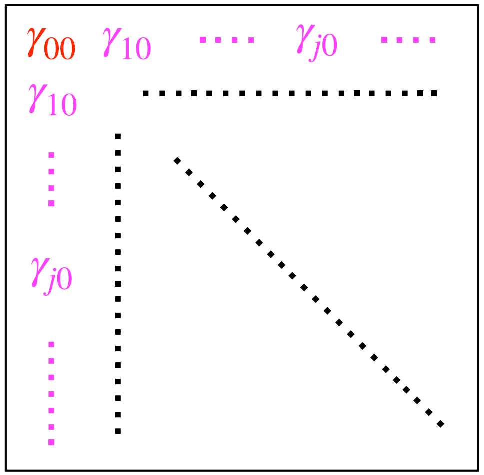
$$\mathbf{P} = \mathbf{I} - 2\mathbf{v}\mathbf{v}^{\dagger}$$

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

A. S. Householder, J. ACM 5, 339 (1958).
S. Sekaran, M. Tsuchiizu, M. Saubanère, and E. Fromager, ***arXiv:2103.04194*** *(2021).*

Householder transformed density matrix embedding





γ *(Lattice representation)*

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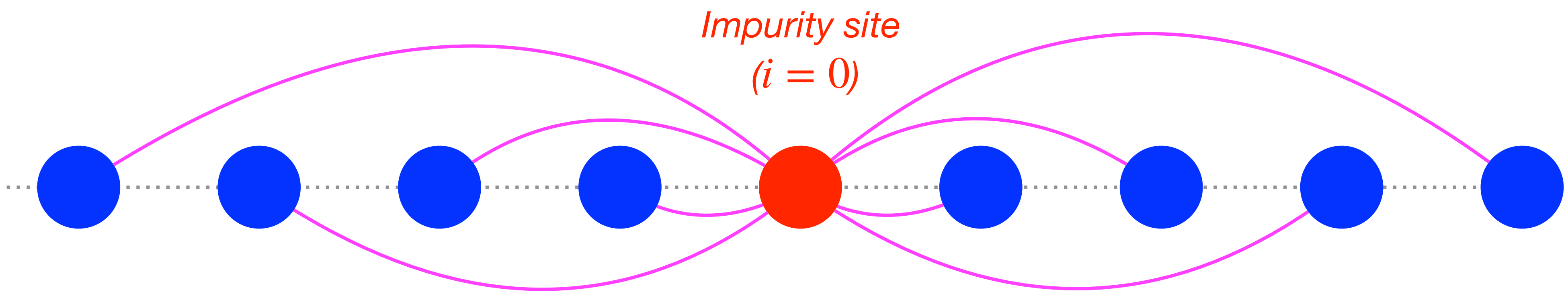

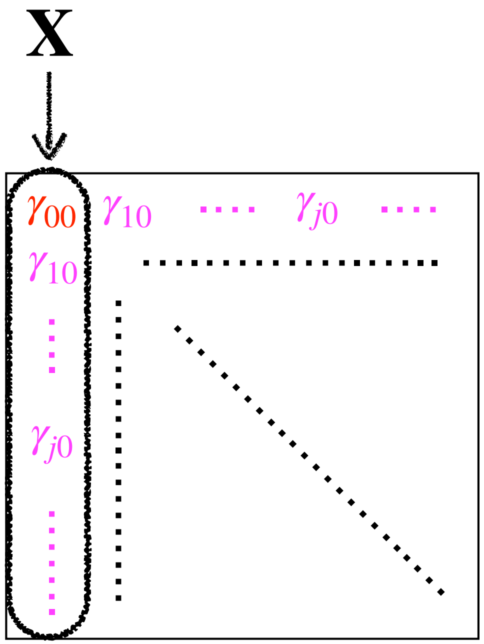

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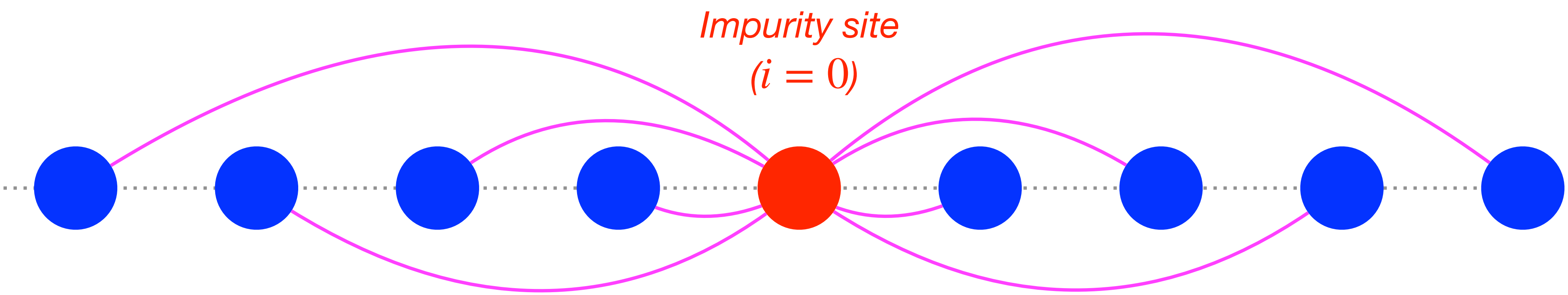
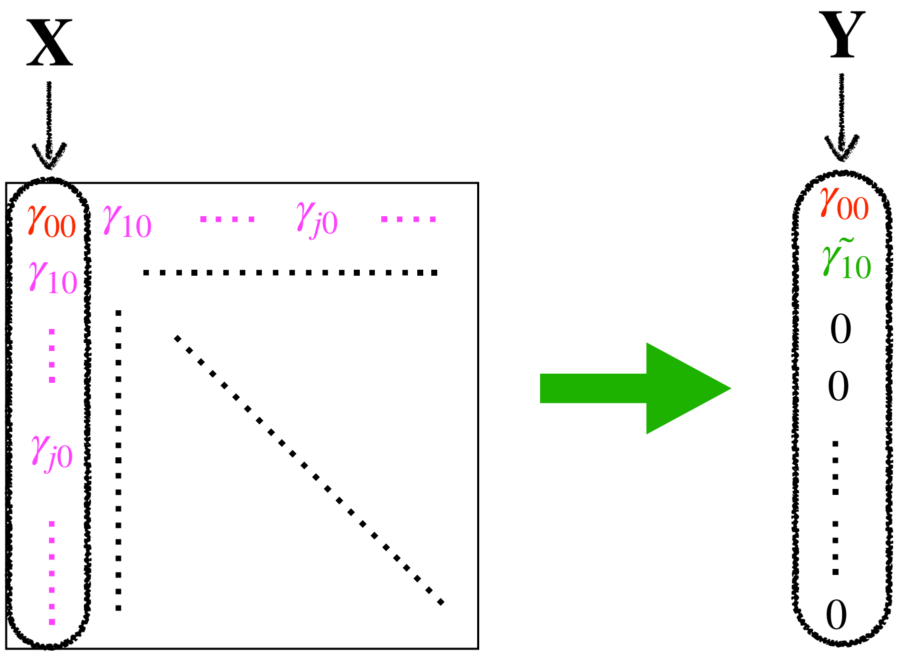
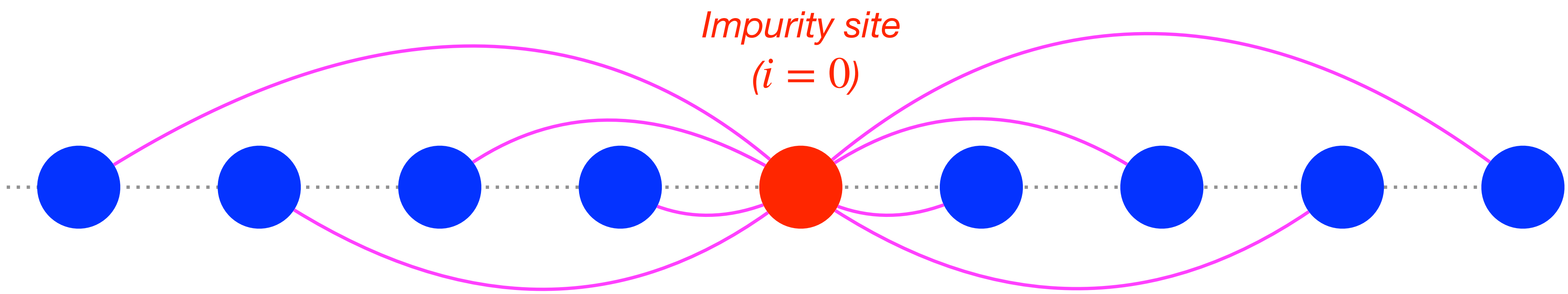


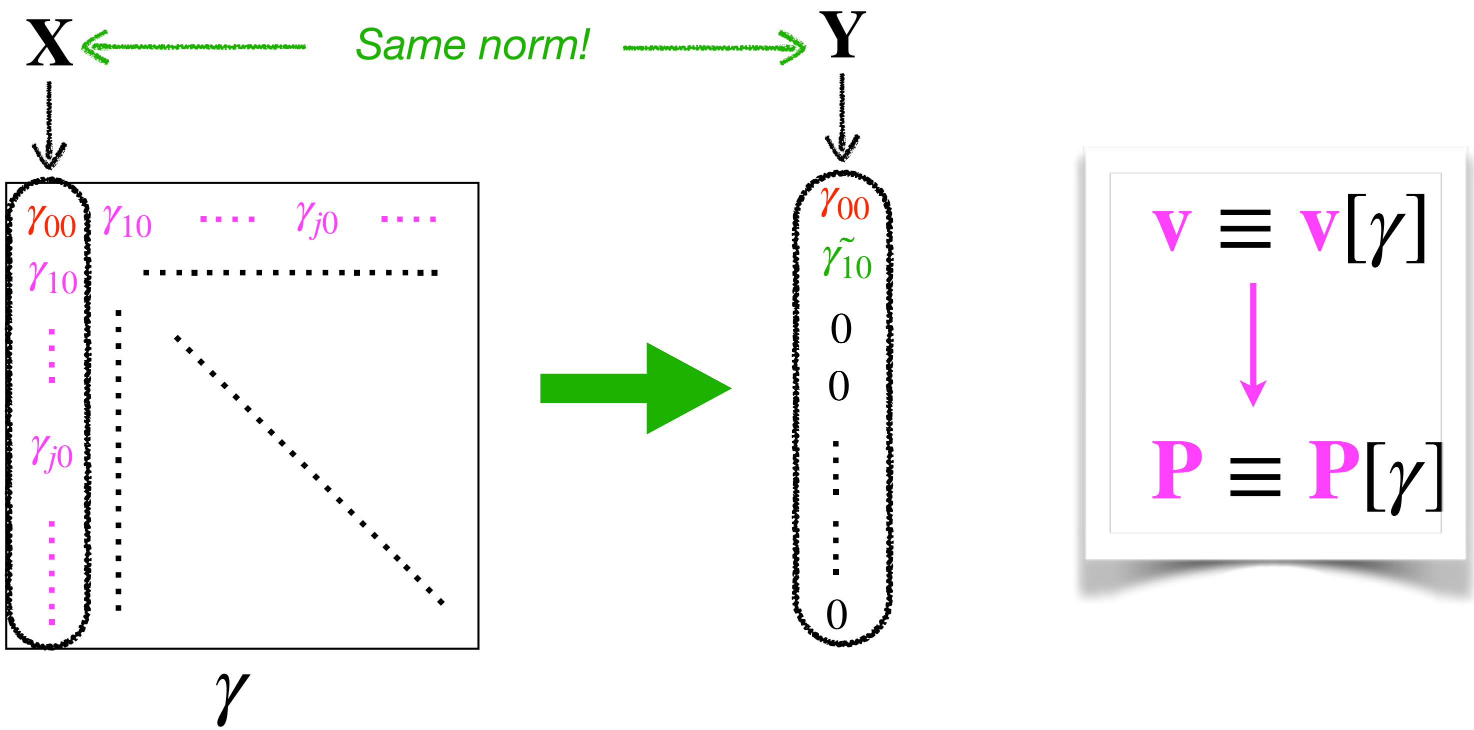


“$\mathbf{X}$ as we would like it to be”

S. Sekaran, M. Tsuchiizu, M. Saubanère, and E. Fromager, ***arXiv:2103.04194*** *(2021).*

Householder transformed density matrix embedding

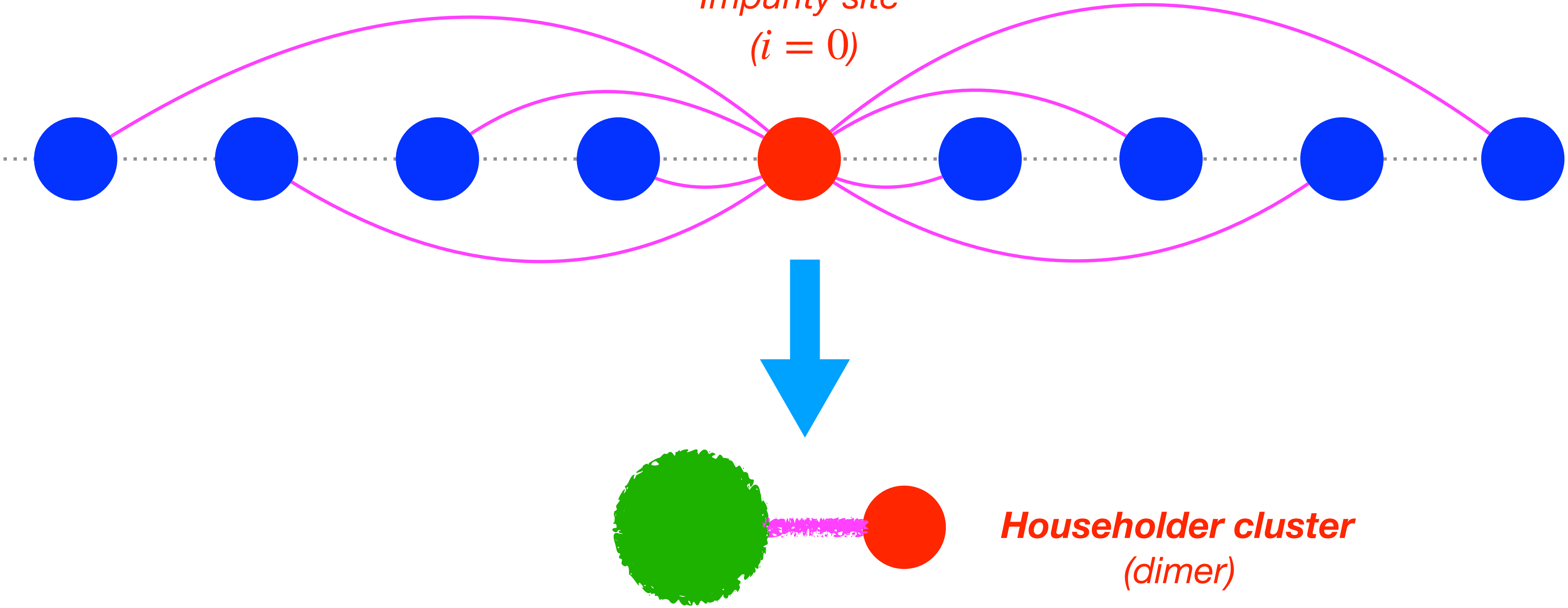




The Householder transformation is an ***explicit functional*** *of the density matrix!*

S. Sekaran, M. Tsuchiizu, M. Saubanère, and E. Fromager, ***arXiv:2103.04194*** *(2021).*

Householder transformed density matrix embedding



"Fermionic mode transformation"

$$\hat{d}^{\dagger}_{\text{bath}} \equiv \hat{d}^{\dagger}_{1\sigma} = \sum_{i \geq 1} P_{1i}\,\hat{c}^{\dagger}_{i\sigma}$$

$$= \hat{c}^{\dagger}_{1\sigma} - 2v_1 \sum_{i \geq 1} v_i\,\hat{c}^{\dagger}_{i\sigma}$$

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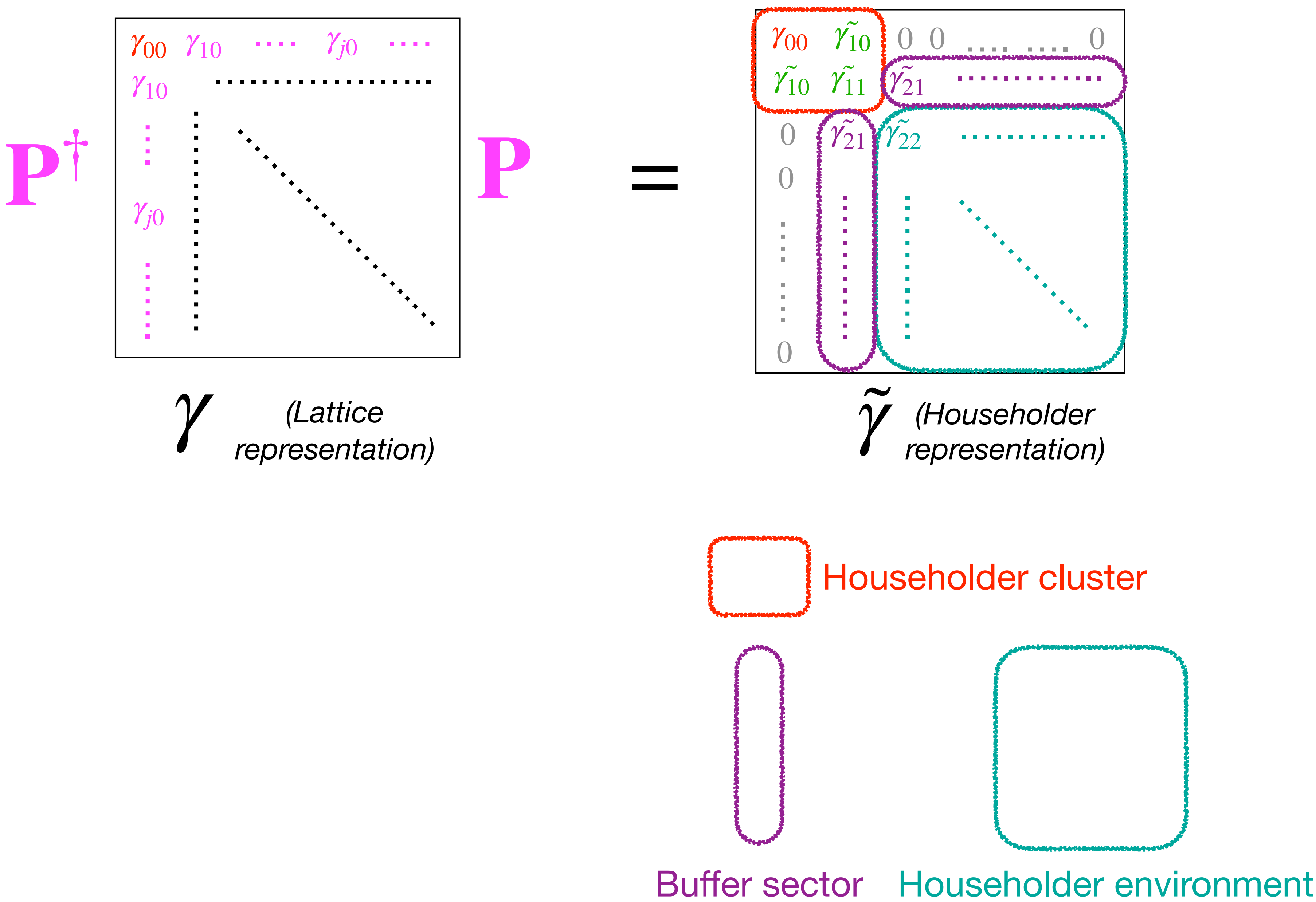
Householder transformed density matrix embedding



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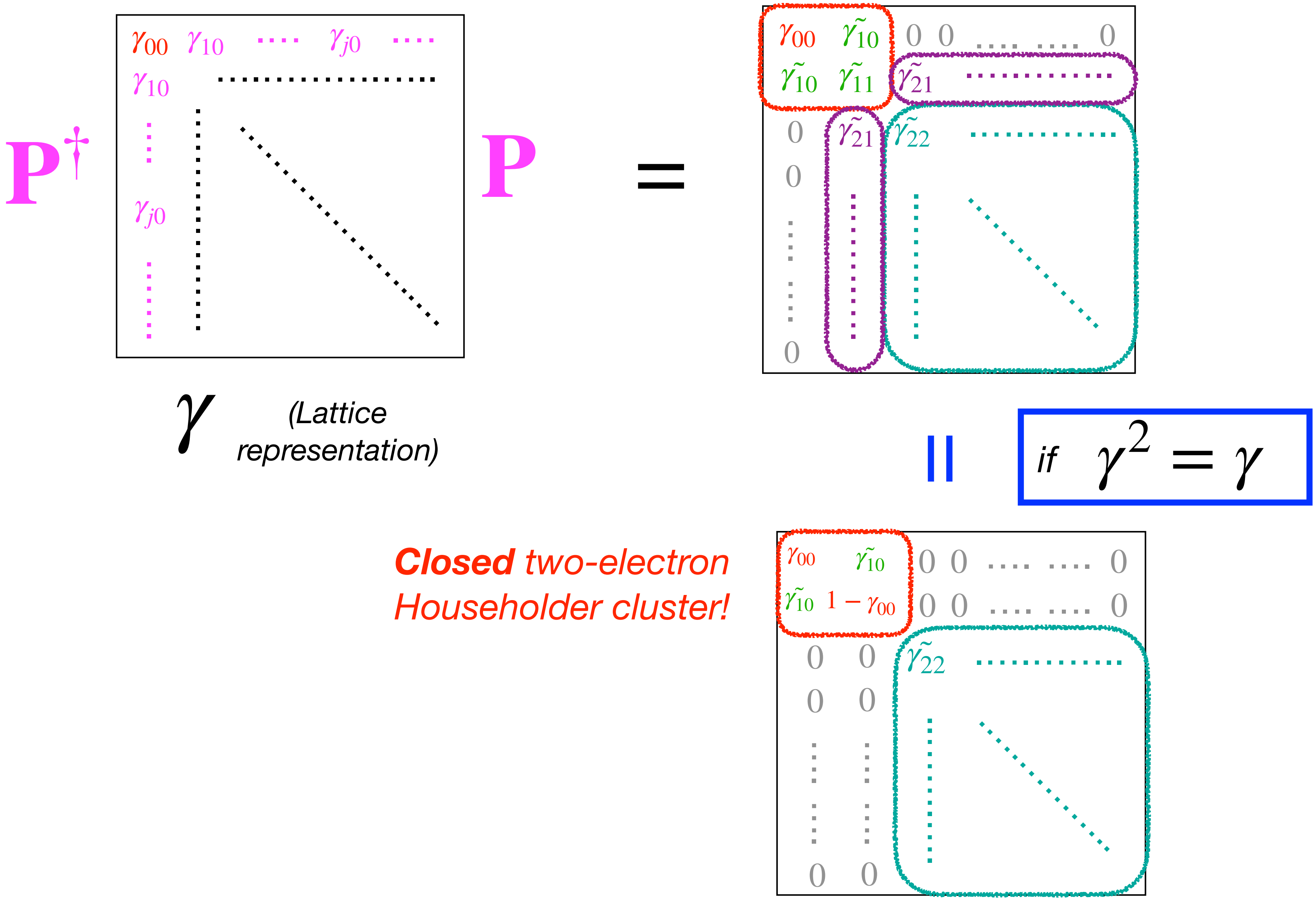

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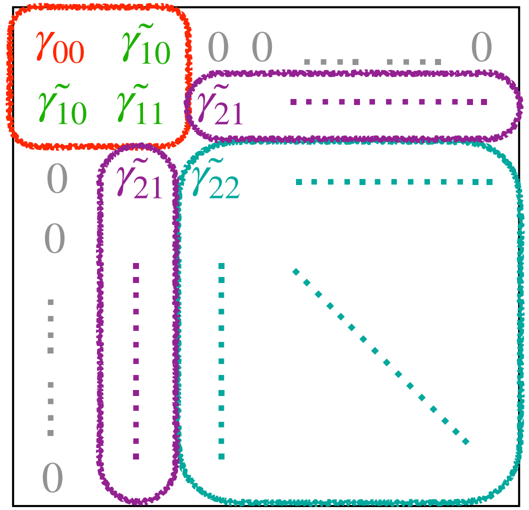
Householder transformed density matrix embedding

The impurity will exchange electrons
with the bath site only



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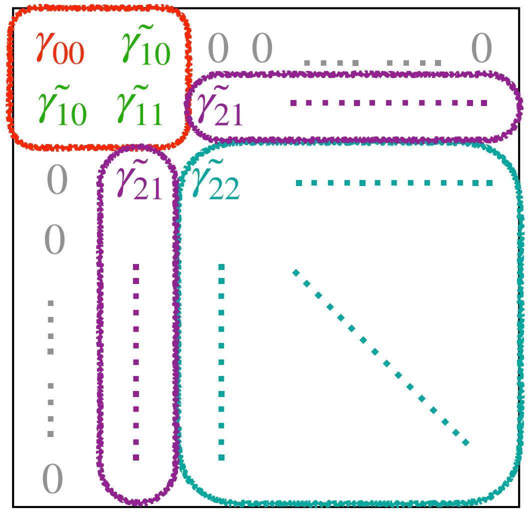
Householder transformed density matrix embedding

Bath state

$$|\Psi\rangle = \sum_{B} |I^B\rangle |B\rangle \sum_{E^B} \lambda_{I^B B E^B} |E^B\rangle$$

Impurity state

Householder environment state



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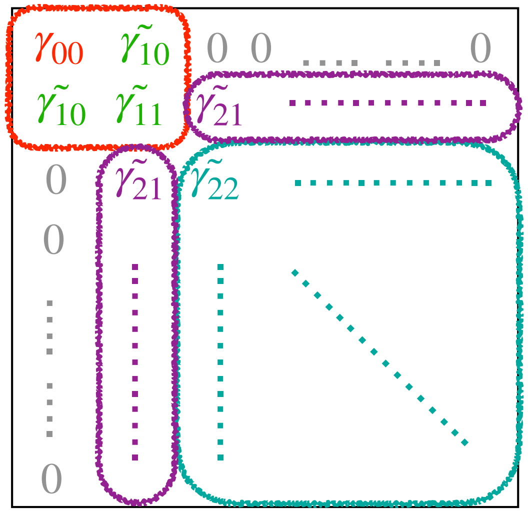
Householder transformed density matrix embedding



Bath state

$$|\Psi\rangle = \sum_{B} |I^B\rangle |B\rangle \sum_{E^B} \lambda_{I^B B E^B} |E^B\rangle$$

Impurity state

Householder environment state

… to be compared with the **Schmidt decomposition** used in DMET*:

$$|\Psi\rangle = \sum_{I} \lambda_I |I\rangle |\mathscr{B}^I\rangle$$

(correlated) ***many-body*** *bath state*

S. Sekaran, M. Tsuchiizu, M. Saubanère, and E. Fromager, ***arXiv:2103.04194*** *(2021).*

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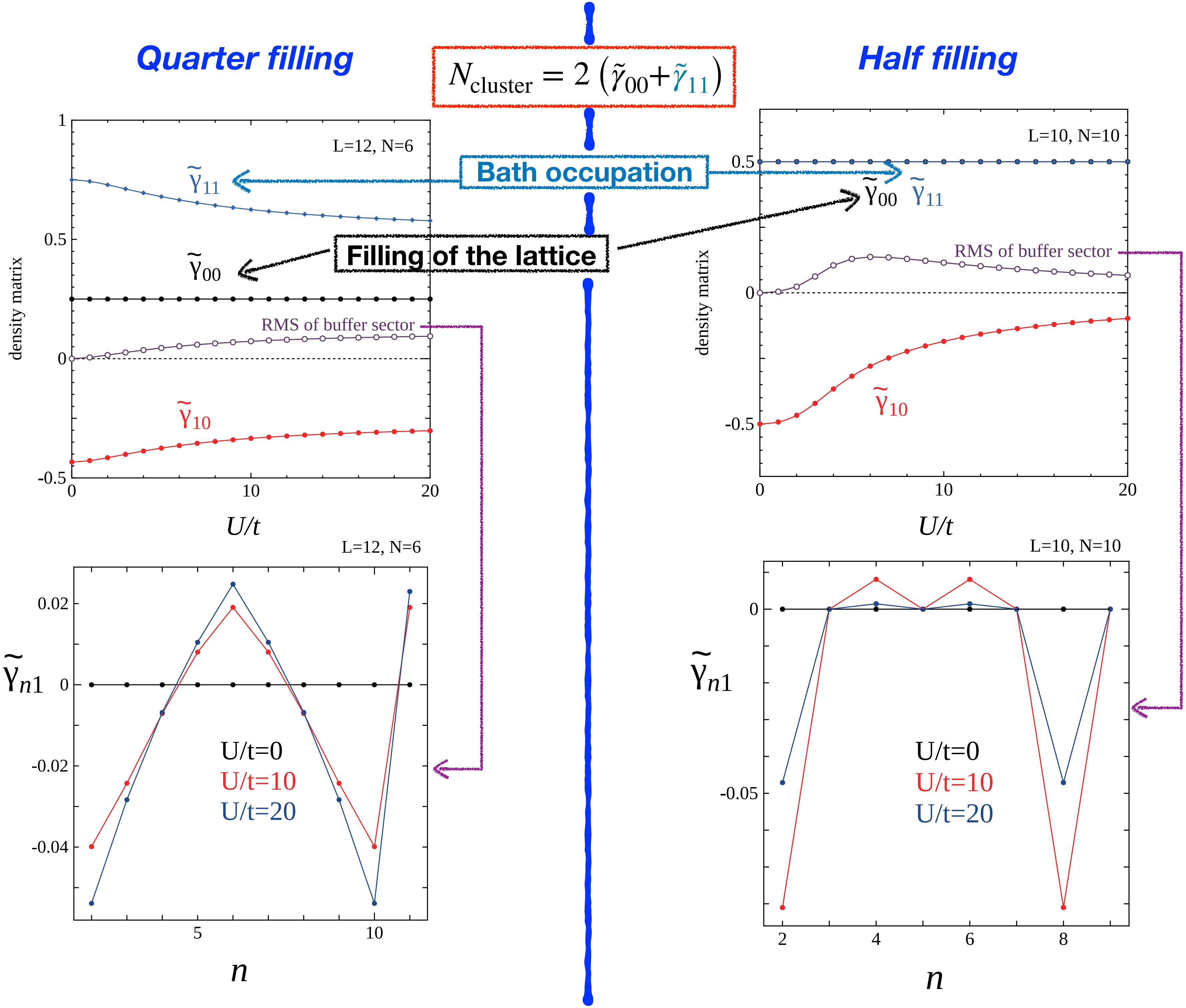

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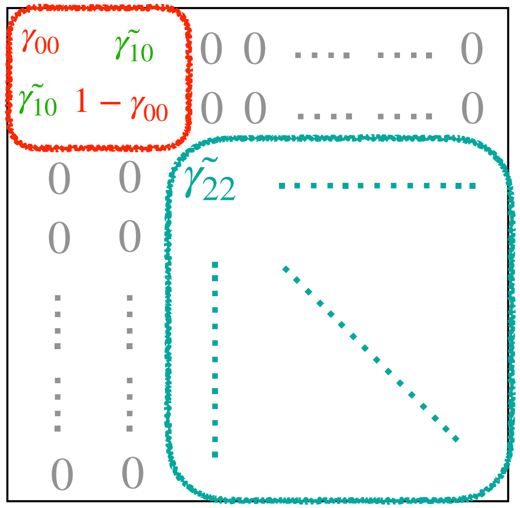
Householder transformed density matrix embedding

$$|\Phi\rangle = \left(\sum_I \lambda_I |I\rangle |B^I\rangle \right) |\Phi_{\text{core}}\rangle$$

$$= |\Phi_{\text{cluster}}\rangle |\Phi_{\text{core}}\rangle$$

if $\gamma^2 = \gamma$



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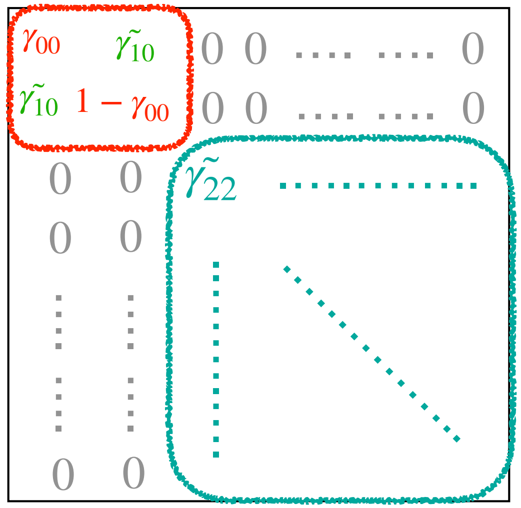
$$= |\Phi_{\text{cluster}}\rangle |\Phi_{\text{core}}\rangle$$

Two-electron cluster's determinant

Householder environment's determinant

if $\gamma^2 = \gamma$



Identical to DMET in this case!

S. Sekaran, M. Tsuchiizu, M. Saubanère, and E. Fromager, ***arXiv:2103.04194*** (2021).

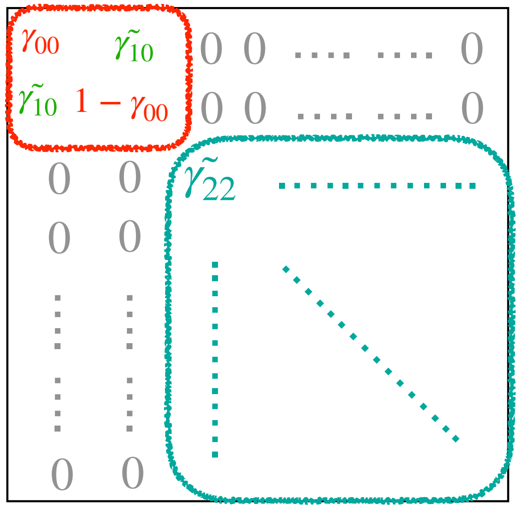
Householder transformed density matrix embedding

$$|\Phi\rangle = \left(\sum_I \lambda_I |I\rangle |B^I\rangle \right) |\Phi_{\text{core}}\rangle$$

$$= |\Phi_{\text{cluster}}\rangle |\Phi_{\text{core}}\rangle$$

if $\gamma^2 = \gamma$

Identical *(but simpler)* to DMET in this case!



The embedding is constructed *analytically* from

$$\gamma_{ij} = \sum_{\kappa}^{\text{occupied}} C_{i\kappa} C_{j\kappa}$$

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Householder transformed density matrix functional embedding theory (Ht-DMFET)

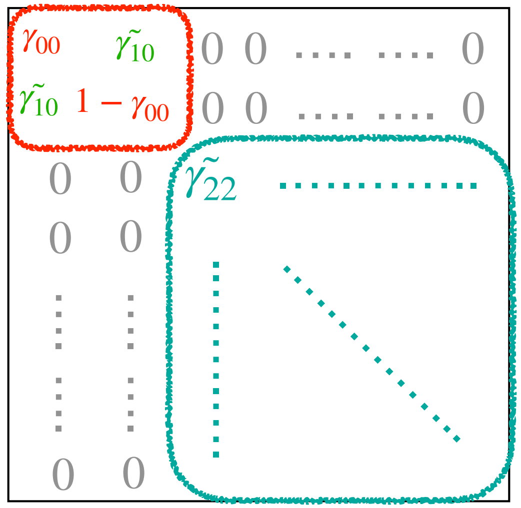
$$|\Phi\rangle = \left(\sum_I \lambda_I |I\rangle |B^I\rangle\right)|\Phi_{\text{core}}\rangle$$

$$= |\Phi_{\text{cluster}}\rangle |\Phi_{\text{core}}\rangle$$

Correlation on top of the non-interacting embedding

$$|\Psi\rangle \approx |\Psi_{\text{cluster}}\rangle |\Phi_{\text{core}}\rangle$$

if $\gamma^2 = \gamma$



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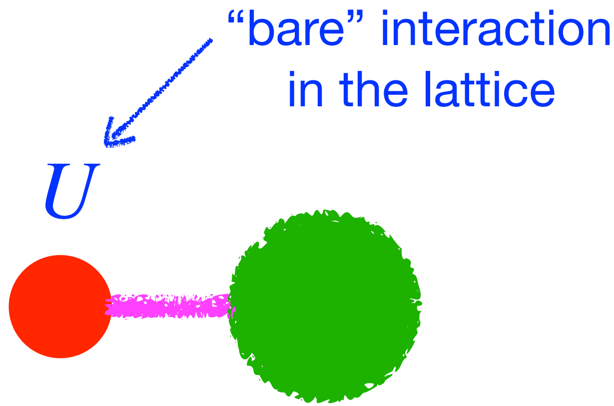
Householder transformed density matrix functional embedding theory (Ht-DMFET)



Non-interacting bath (NIB) approximation

Correlation on top of the non-interacting embedding

$$|\Psi\rangle \approx |\Psi_{\text{cluster}}\rangle|\Phi_{\text{core}}\rangle$$

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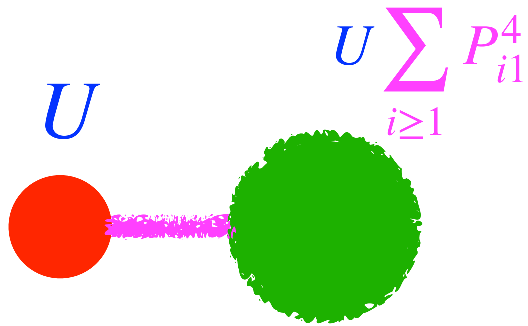
Householder transformed density matrix functional embedding theory (Ht-DMFET)



Interacting bath (IB) approximation

Correlation on top of the non-interacting embedding

$$|\Psi\rangle \approx |\Psi_{\text{cluster}}\rangle |\Phi_{\text{core}}\rangle$$

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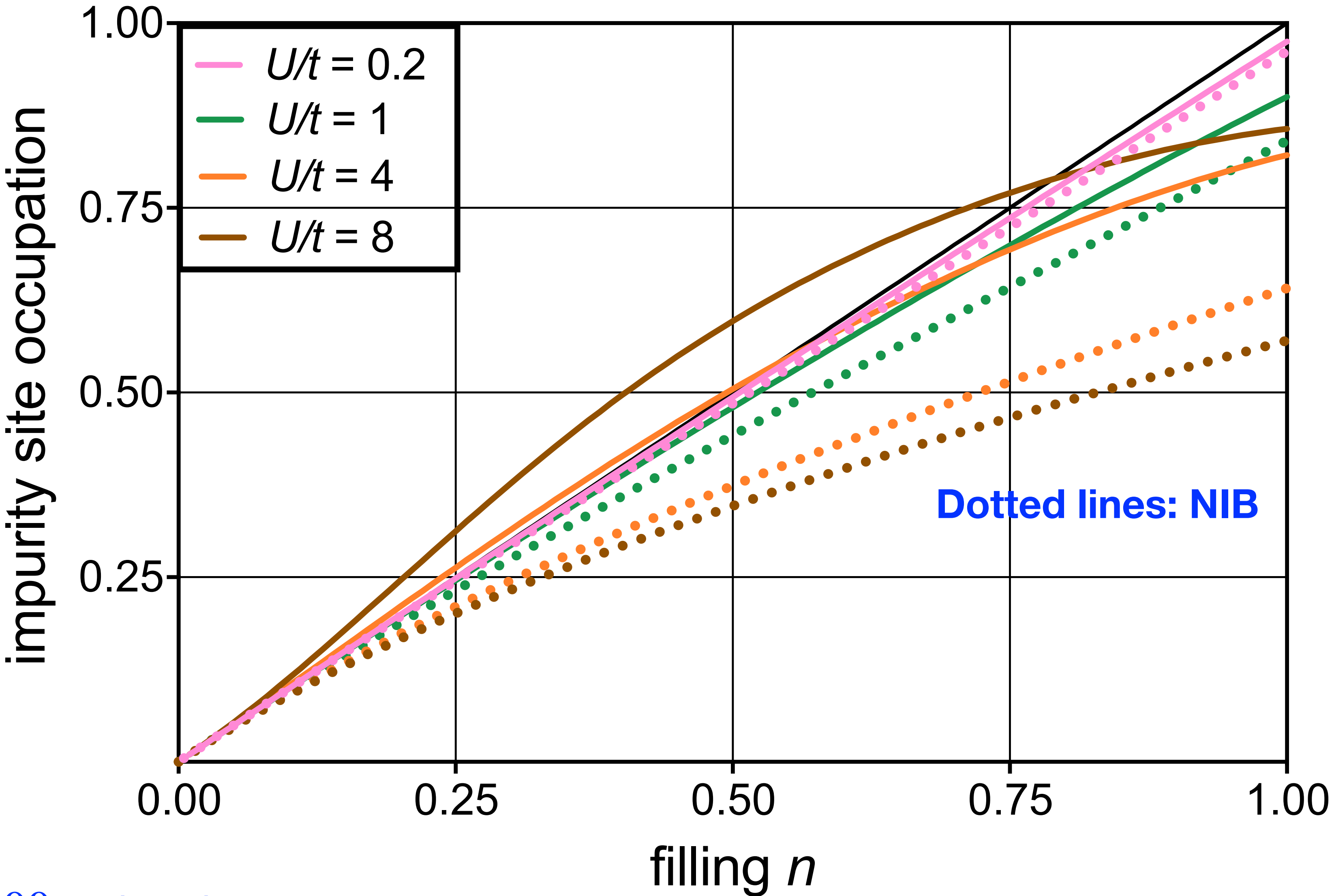
Householder transforming the Hamiltonian and projecting onto the cluster



$L = 400$ lattice sites

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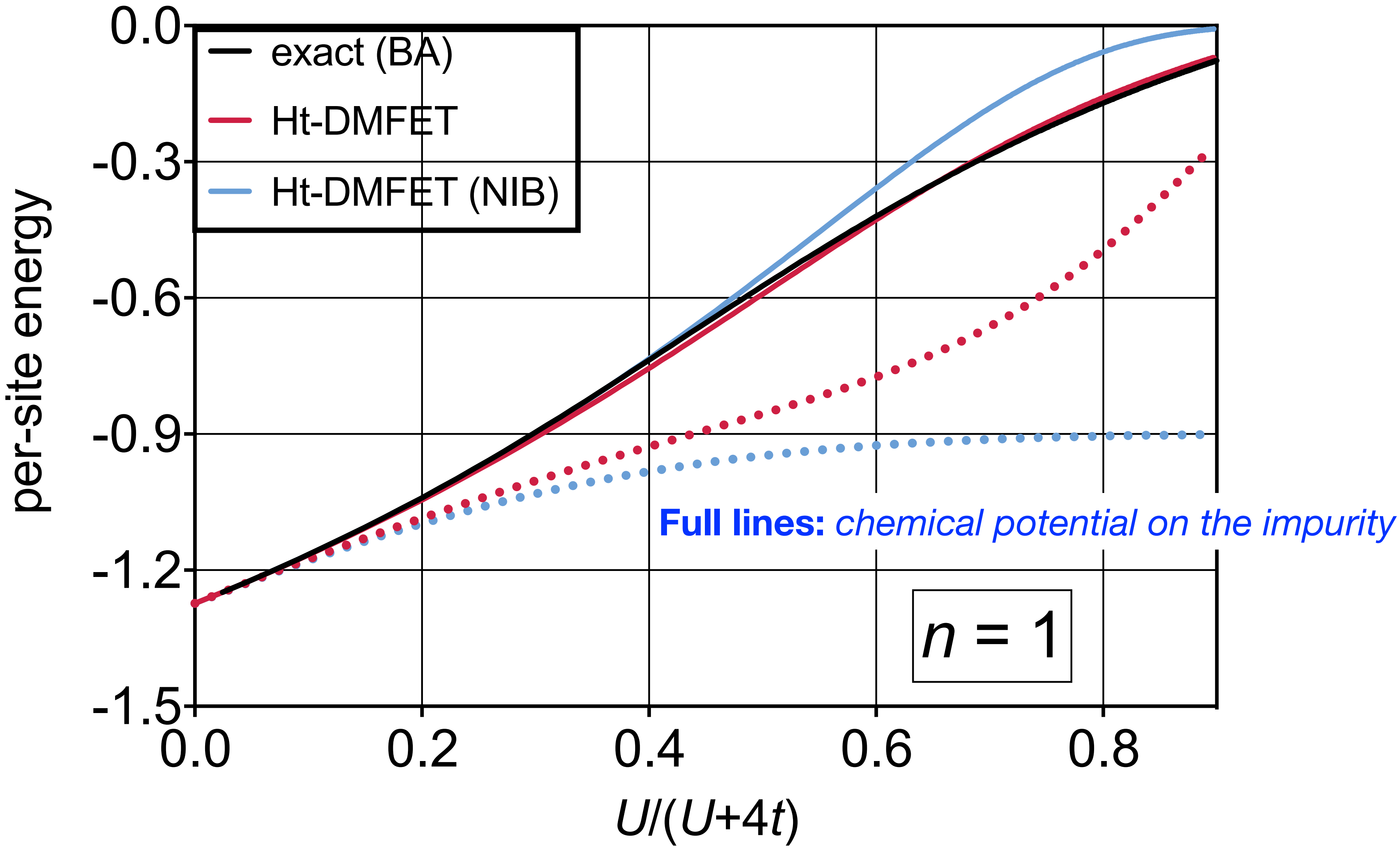
Ht-DMFET per-site energies at half-filling



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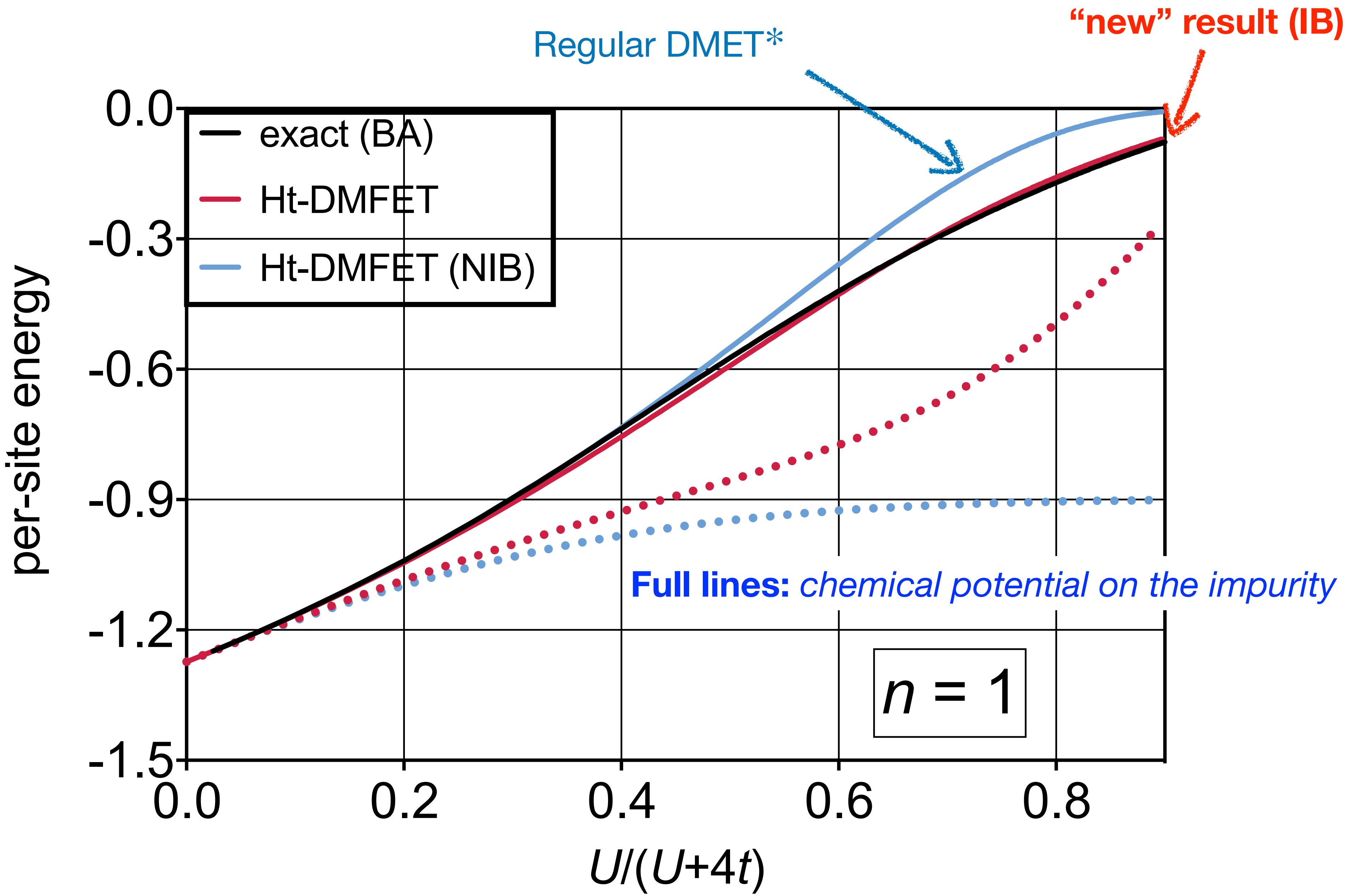

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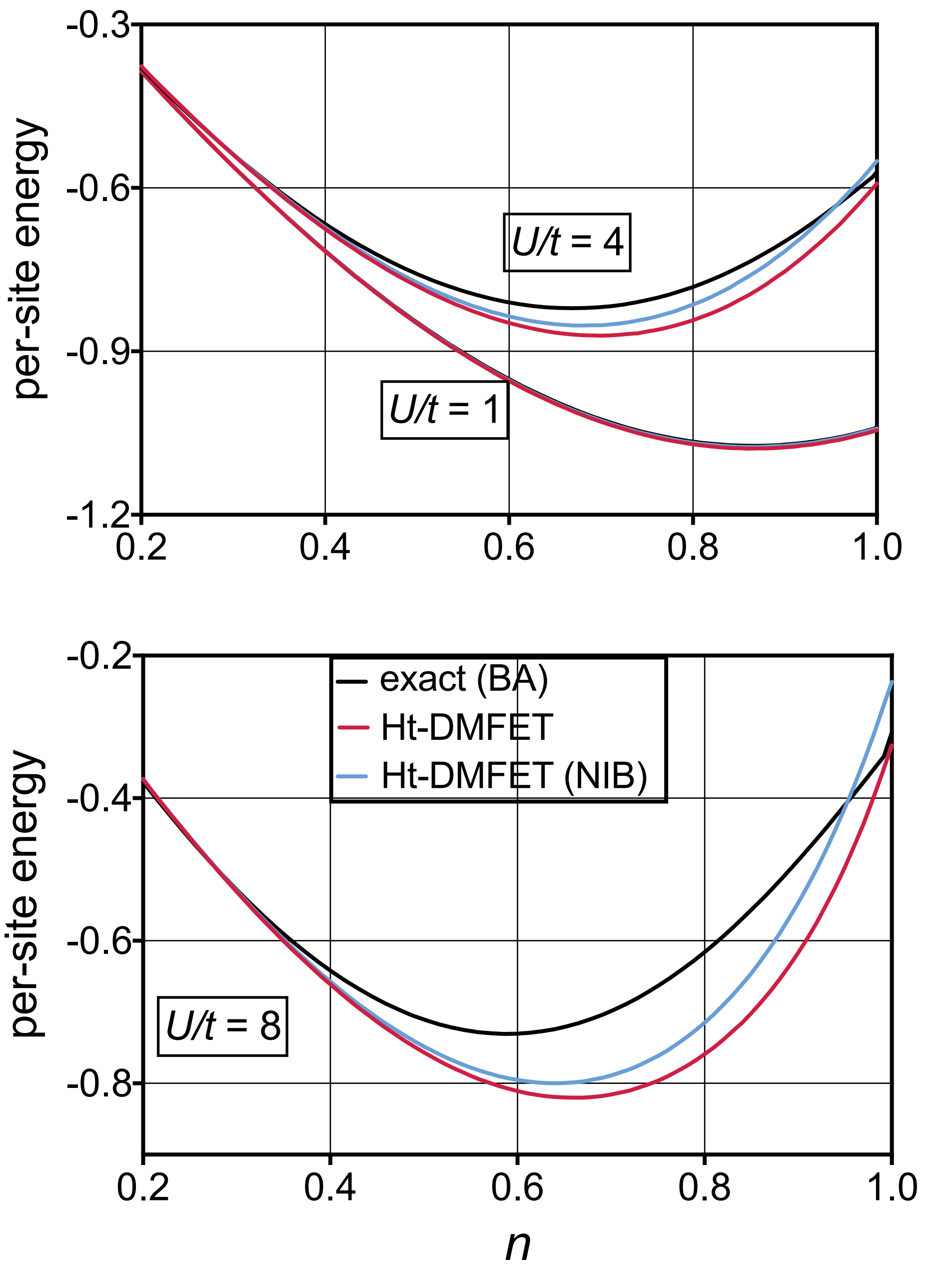
Ht-DMFET per-site energies away from half-filling ($n < 1$)



$L = 400$ lattice sites

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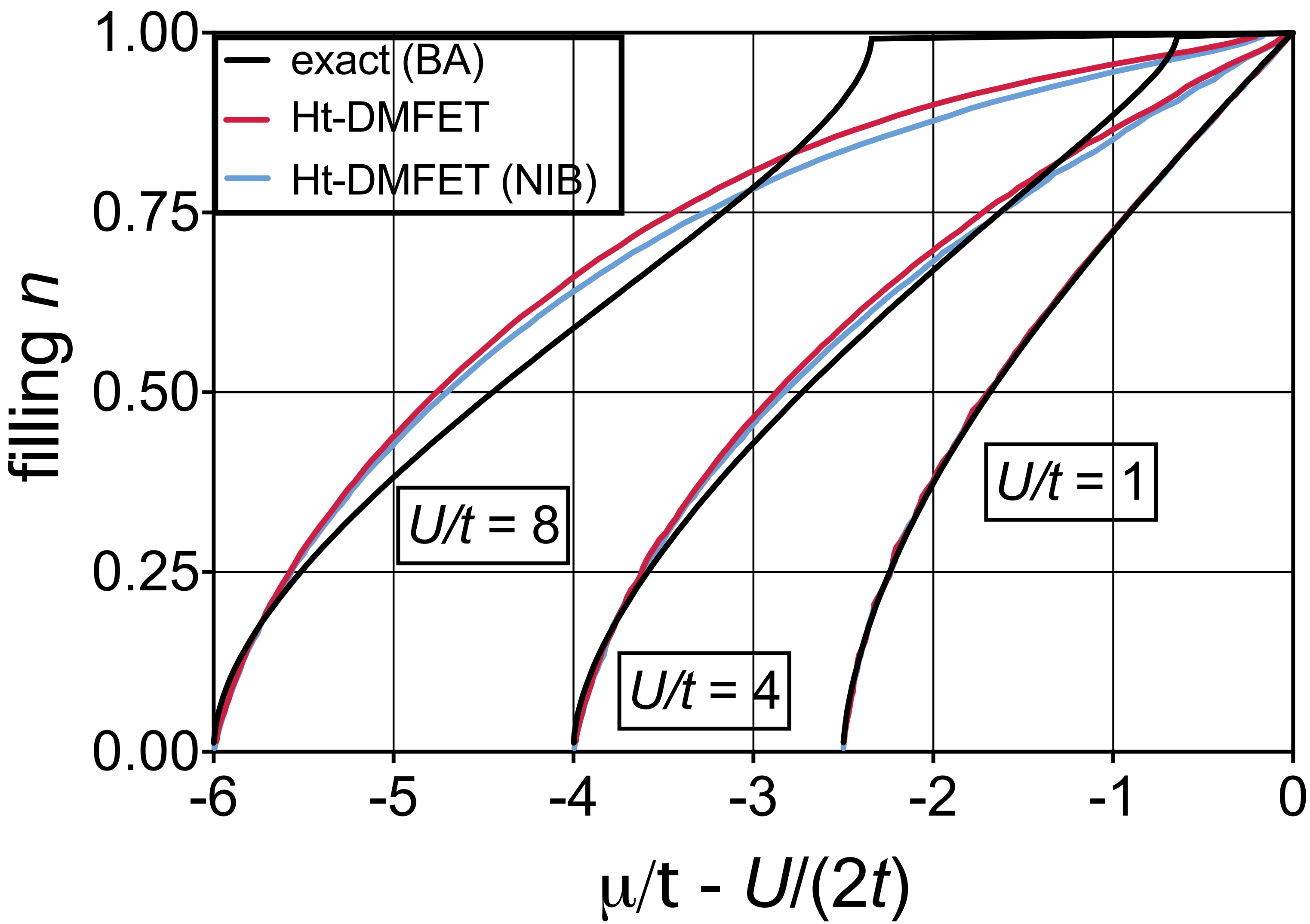
Density-driven Mott-Hubbard transition



$L = 400$ lattice sites

S. Sekaran, M. Tsuchiizu, M. Saubanère, and E. Fromager, ***arXiv:2103.04194*** *(2021).*

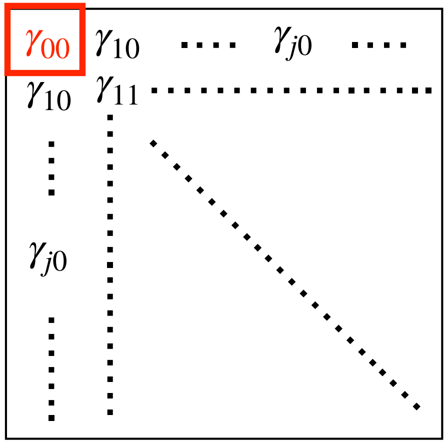
Looking into the near future: Ht-DMFET in Quantum Chemistry

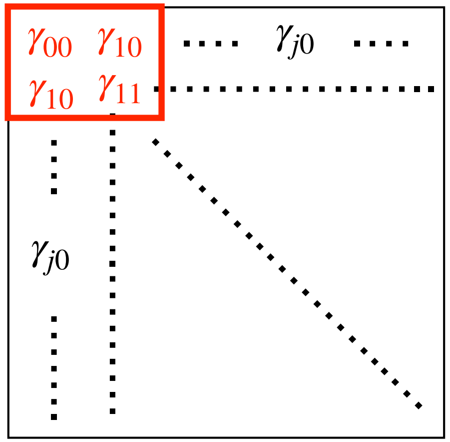
Looking into the near future: Ht-DMFET in Quantum Chemistry

- We currently work on a *multiple-impurity extension* of the theory:

$$\mathbf{P} = \mathbf{I} - 2\mathbf{v}\mathbf{v}^{\dagger} \quad \Rightarrow \quad \mathbf{P} = \mathbf{I} - 2\mathbf{V}\left[\mathbf{V}^{\dagger}\mathbf{V}\right]^{-1}\mathbf{V}^{\dagger}$$

Block Householder** transformation**





F. Rotella and I. Zambettakis, Appl. Math. Lett.* ***12**, 29 (1999).*

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Looking into the near future: Ht-DMFET in Quantum Chemistry

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$$P_{ij}$$

Discrete Householder transformation

Continuous Householder transformation (?)

$$P \equiv \mathscr{P}(\mathbf{r},\mathbf{r}') = \delta(\mathbf{r}-\mathbf{r}') - 2\int_{\mathscr{F}} d\mathbf{r}_1 \int_{\mathscr{F}} d\mathbf{r}_2\, V(\mathbf{r},\mathbf{r}_1)\left[V^{\dagger}V\right]^{-1}(\mathbf{r}_1,\mathbf{r}_2)V(\mathbf{r}',\mathbf{r}_2)$$

Looking into the near future: Ht-DMFET in Quantum Chemistry

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Localised orbitals

$$\int_{\mathscr{F}} d\mathbf{r}_1 \int_{\mathscr{F}} d\mathbf{r}_2 \equiv \int d\mathbf{r}_1 \int d\mathbf{r}_2 \sum_{i\in\mathscr{F}} \phi_i(\mathbf{r}_1)\phi_i(\mathbf{r}_2) \times$$

Integration over a fragment

$$P \equiv \mathscr{P}(\mathbf{r},\mathbf{r}') = \delta(\mathbf{r}-\mathbf{r}') - 2\int_{\mathscr{F}} d\mathbf{r}_1 \int_{\mathscr{F}} d\mathbf{r}_2\, V(\mathbf{r},\mathbf{r}_1)\left[V^\dagger V\right]^{-1}(\mathbf{r}_1,\mathbf{r}_2)V(\mathbf{r}',\mathbf{r}_2)$$

Looking into the near future: Ht-DMFET in Quantum Chemistry

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Semi-local DFA treatment

$$E_c \approx E_{c,\mathscr{F}}^{\mathrm{Ht-DMFET}} + \int d\mathbf{r}_1 \int d\mathbf{r}_2 \left(\delta(\mathbf{r}_1 - \mathbf{r}_2) - \sum_{i \in \mathscr{F}} \phi_i(\mathbf{r}_1)\phi_i(\mathbf{r}_2) \right) n(\mathbf{r}_2)\, \varepsilon_c\big(n(\mathbf{r}_1), |\nabla n(\mathbf{r}_1)|, \ldots\big)$$

*… In the spirit of domain separated DFT**

M. A. Mosquera, L. O. Jones, C. H. Borca, M. A. Ratner, and G. C. Schatz, J. Phys. Chem. A **123, 4785 (2019).*

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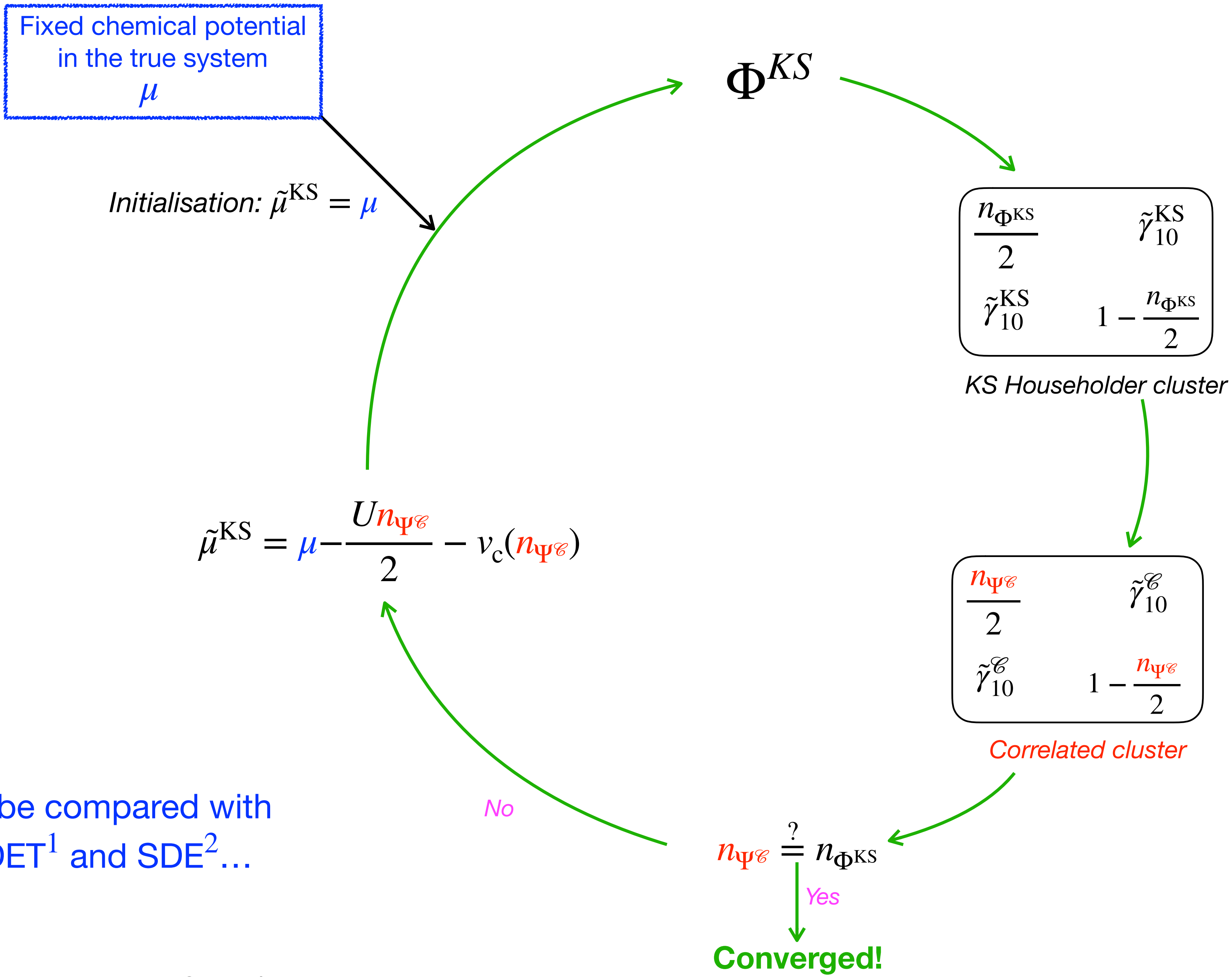
LabEx CSC (ANR-10-LABX-0026-CSC)

CoLab ANR project

Householder density-functional embedding theory (H-DFET)



To be compared with
DET[1] and SDE[2]...

S. Sekaran, M. Tsuchiizu, M. Saubanère, and E. Fromager, in preparation.

[1] *W. Bulik, G. E. Scuseria, and J. Dukelsky, Phys. Rev. B* ***89****, 035140 (2014).*

[2] *U. Mordovina, T. E. Reinhard, I. Theophilou, H. Appel, and A. Rubio, J. Chem. Theory Comput.* ***15****, 5209 (2019).*

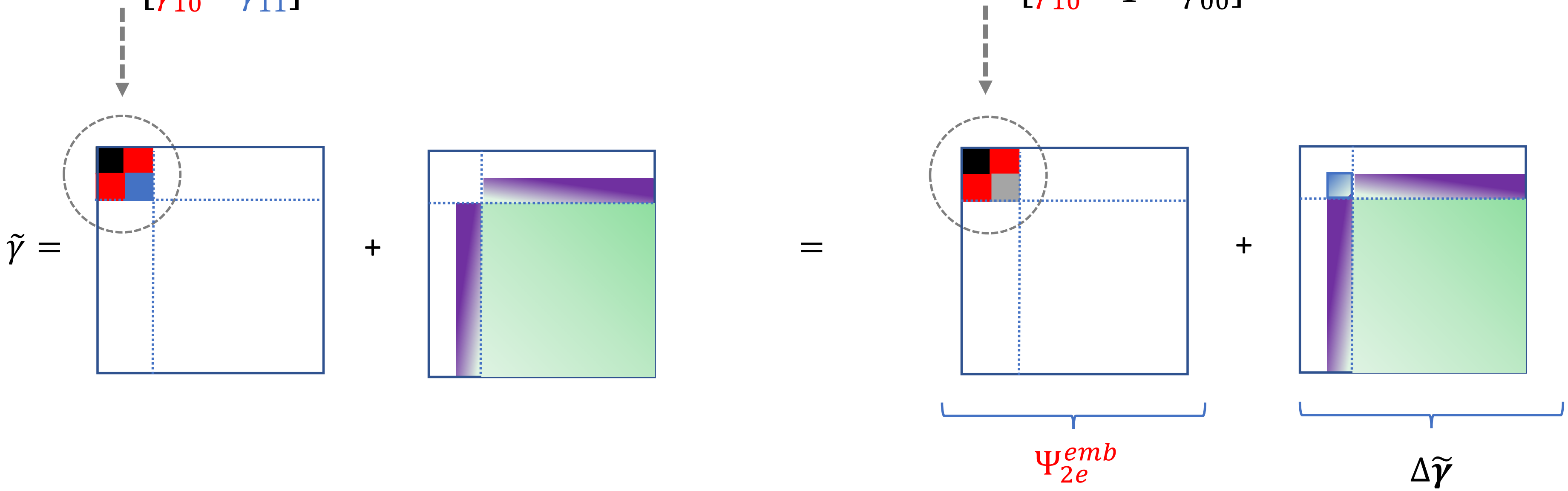
Formally exact Ht-DMFET?

How to?

We may close the cluster while preserving the exact off-diagonal elements of the 1RDM.

***Open** cluster:* $\begin{bmatrix} \tilde{\gamma}_{00} & \tilde{\gamma}_{10} \\ \tilde{\gamma}_{10} & \tilde{\gamma}_{11} \end{bmatrix} = \tilde{\boldsymbol{\gamma}}^{\mathcal{C}}$

***Two-electron** cluster:* $\begin{bmatrix} \tilde{\gamma}_{00} & \tilde{\gamma}_{10} \\ \tilde{\gamma}_{10} & 1 - \tilde{\gamma}_{00} \end{bmatrix} \xleftarrow{???} \Psi_{2e}^{emb}$



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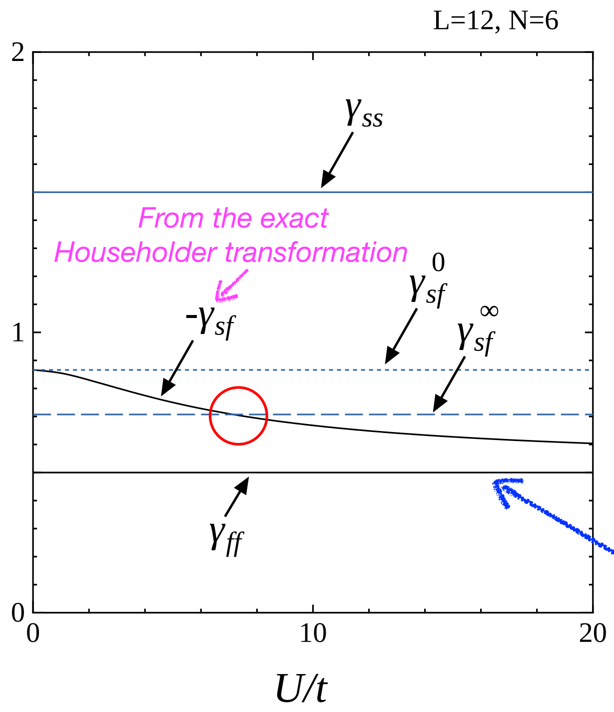
v-representability issues

two-electron cluster



Pure-state Anderson-v-representability condition: $\boxed{\gamma_{sf}^{\infty} \leq |\gamma_{sf}| \leq \gamma_{sf}^{0}}$, where

$$\gamma_{sf}^{0} = \sqrt{\gamma_{ff}\,(2 - \gamma_{ff})}$$

$$\gamma_{sf}^{\infty} = \sqrt{2\gamma_{ff}\,(1 - \gamma_{ff})} \quad \text{for } \gamma_{ff} \leq 1.$$

Impurity occupation $\equiv N/L$

Ground-state wave function

$$\gamma_{ff} \equiv \sum_{\sigma=\uparrow,\downarrow} \langle \Psi_{2e}^{emb} | \hat{c}_{f\sigma}^{\dagger} \hat{c}_{f\sigma} | \Psi_{2e}^{emb} \rangle$$

and

$$\gamma_{sf} \equiv \sum_{\sigma=\uparrow,\downarrow} \langle \Psi_{2e}^{emb} | \hat{c}_{s\sigma}^{\dagger} \hat{c}_{f\sigma} | \Psi_{2e}^{emb} \rangle$$

W. Töws and G. Pastor, Phys. Rev. B 83, 235101 (2011).

S. Sekaran, M. Tsuchiizu, M. Saubanère, and E. Fromager, in preparation.