Exploiting the Semidefinite Programming Formulations on the Variational Calculation of Second-Order Reduced Density Matrix of Atoms and Molecules

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Outline of the Talk

- How to obtain a lower bound for the ground state energy of fermionic systems by 2nd-order reduced density matrices?
- Historical notes, N-representability conditions
- Semidefinite Programming (SDP)
- Primal SDP formulation and dual SDP formulation
- Theoretical comparison on computational complexity with RRSDP (Mazziotti)
- Numerical Results

Today 4:15-4:45 Maho Nakata, “The Reduced Density Matrix Method: Applications of $T2'$ N-representability Conditions and Development of Highly Accurate Solver”

Variational Calculation on 2nd-Order RDM

- determine the ground-state (energy) of a fermionic system by a variational calculation
- variables are the Second-Order Reduced Density Matrices (2-RDMs)
- need to impose the so-called necessary N-representability conditions
- since the N-representability conditions are only necessary (and not sufficient), we can only obtain a lower bound for the ground state energy
- it can be formulated mathematically as an Semidefinite Programming Problem (SDP)
- SDPs can be solved efficiently by Interior-Point Methods
- provides an extremely good approximation, but there is a serious limit on the size of the system in general orbital basis

Incomplete List on the 2-RDM Computation

1940	Husimi 1955 Löwdin 1955 Mayer	RDM
1960	Coulson	2-RDM
1963	Coleman	N-representability conditions
1964	Garrod-Percus	G condition
1960's-1970's	Kijewski, Garrod-Mihailović-Rosina, Garrod-Fusco, Erdahl	
1975	Mihailović-Rosina	nucleon systems
2001	Nakata-Nakatsuji-Ehara-Fukuda-Nakata-Fujisawa (JCP **114** 8282)	
2002	Nakata-Ehara-Nakatsuji (JCP **116** 5432)	potential energy surface
2002	Mazziotti (PRA **65** 062511)	
2004	Zhao-Braams-Fukuda-Overton-Percus (JCP **120** 2095)	$T1, T2$
2004	Mazziotti (PRL **93** 213001)	RRSDP
2006	Cancès-Stoltz-Lewin (JCP **125** 064101)	dual
2007	Braams-Percus-Zhao (ACP vol. **134**)	$T2'$ condition
2006,2007	Mazziotti (PRA **74** 32501, ACP vol. **134**)	$\bar{T}2$ condition
2008	Nakata-Braams-Fujisawa-Fukuda-Percus-Yamashita-Zhao (JCP **128** 164113)	

1st- and 2nd-Order RDMs

- variational calculation which involves only the 1-RDM

$$\gamma^{i_1}_{j_1} = \langle \Psi | a^\dagger_{i_1} a_{j_1} | \Psi \rangle$$

and the 2-RDM

$$\Gamma^{i_1 i_2}_{j_1 j_2} = \frac{1}{2} \langle \Psi | a^\dagger_{i_1} a^\dagger_{i_2} a_{j_2} a_{j_1} | \Psi \rangle$$

- we impose some conditions on the 1-RDM and 2-RDM in order to be N-representable,
that is, there must exists an anti-symmetric wavefunction

$$\Psi(\ldots, i, \ldots, j, \ldots) = -\Psi(\ldots, j, \ldots, i, \ldots)$$

which results in the 1-RDM and 2-RDM above

Known N-representability conditions

P: $2\Gamma^{i_1 i_2}_{j_1 j_2}$ — Coleman 1963

Q: $(\delta^{i_1}_{j_1}\delta^{i_2}_{j_2} - \delta^{i_1}_{j_2}\delta^{i_2}_{j_1}) - (\delta^{i_1}_{j_1}\gamma^{i_2}_{j_2} + \delta^{i_2}_{j_2}\gamma^{i_1}_{j_1}) + (\delta^{i_1}_{j_2}\gamma^{i_2}_{j_1} + \delta^{i_2}_{j_1}\gamma^{i_1}_{j_2}) + 2\Gamma^{i_1 i_2}_{j_1 j_2}$ — Coleman 1963

G: $\delta^{i_2}_{j_2}\gamma^{i_1}_{j_1} - 2\Gamma^{i_1 j_2}_{j_1 i_2}$ — Garrod-Percus 1964

kth-order approximation: — Erdahl-Jin 2000

$T1$: $\mathcal{A}[i_1, i_2, i_3]\mathcal{A}[j_1, j_2, j_3]\left(\frac{1}{6}\delta^{i_1}_{j_1}\delta^{i_2}_{j_2}\delta^{i_3}_{j_3} - \frac{1}{2}\delta^{i_1}_{j_1}\delta^{i_2}_{j_2}\gamma^{i_3}_{j_3} + \frac{1}{2}\delta^{i_1}_{j_1}\Gamma^{i_2 i_3}_{j_2 j_3}\right)$ — Erdahl 1978, Zhao *et al.* 2004

$T2$: $\mathcal{A}[i_2, i_3]\mathcal{A}[j_2, j_3]\left(\frac{1}{2}\delta^{i_2}_{j_2}\delta^{i_3}_{j_3}\gamma^{i_1}_{j_1} + \frac{1}{2}\delta^{i_1}_{j_1}\Gamma^{j_2 j_3}_{i_2 i_3} - 2\delta^{i_2}_{j_2}\Gamma^{i_1 j_3}_{j_1 i_3}\right)$ — Erdahl 1978, Zhao *et al.* 2004

$T2'$: $\begin{pmatrix} T2 & X \\ X^\dagger & \gamma \end{pmatrix}$ where $X^k_{i_1 i_2 i_3} = \Gamma^{i_1 k}_{i_2 i_3}$ — Erdahl 1978, Braams-Percus-Zhao 2007, Mazziotti 2006,2007

$\mathcal{A}[i,j,k]f(i,j,k) = f(i,j,k) - f(i,k,j) - f(j,i,k) + f(j,k,i) + f(k,i,j) - f(k,j,i)$

Kronecker's delta δ^i_j

N-representability Conditions (Open Problem)

- complete set of N-representability conditions on the 1-RDM:

$$\boldsymbol{I} \succeq \gamma \quad \gamma \succeq \mathbf{0}$$

- for the 2-RDM is an extremely difficult problem
- The Diagonal Problem: determine all the N-representability conditions for the diagonal elements of 2-RDM is NP-hard

  cf. Deza-Laurent, *Geometry of Cuts and Metrics*, Springer-Verlag, 1997
- the decision problem: If a given 2-RDM is N-representable is Quantum Merlin-Arthur complete (QMA-complete) $\Rightarrow$ NP-hard

  cf. Y. Liu, M. Christandl, F. Verstraete, Phys. Rev. Lett. **98** 110503 (2007)

Variational Calculation by SDP

- impose only the known N-representability conditions such as P, Q, G, $T1$, $T2'$ conditions and perform the variational calculation on 1-RDM and 2-RDM
- computes a lower bound for the ground state energy and an approximate 1- and 2-RDMs

$$\left\{\begin{array}{ll} \text{minimize} & \text{tr}(H_1\gamma) + \text{tr}(H_2\Gamma) \\ \text{subject to} & P, Q, G, T1, T2' \text{ conditions} \end{array}\right.$$

$\Rightarrow$ $\boxed{\text{Semidefinite Programming Problem}}$

Optimization and SDP

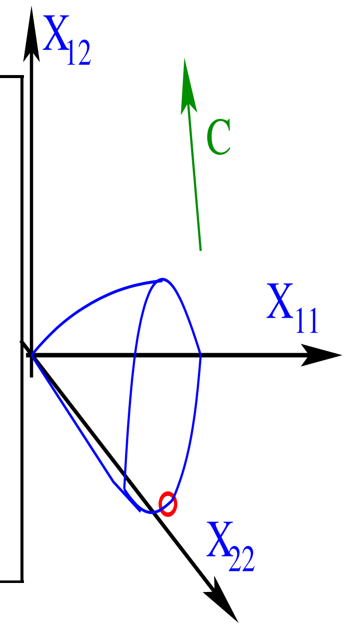
- Optimization or Mathematical Programming

  $\Rightarrow$ Develop efficient algorithms in theory and in practice to solve optimization problems (generally involving finite dimensional vectors, matrices or graphs)

- Semidefinite Programming Problem (SDP)
  - Linear Matrix Inequality (LMI) in system and control theory
  - natural extension of Linear Programming (LP)
  - can be solved efficiently by Interior-Point Methods
  - powerful mathematical model which can efficiently approximate problems which are essentially quadratic

Semidefinite Program (SDP)

$$
\begin{array}{lll}
\texttt{Primal} & \left\{ \begin{array}{ll} \textbf{minimize} & \sum_{i=1}^{\ell} \mathrm{tr}(\boldsymbol{C}_i \boldsymbol{X}_i) \\ \textbf{subject to} & \sum_{i=1}^{\ell} \mathrm{tr}(\boldsymbol{A}_{ip} \boldsymbol{X}_i) = b_p \quad (1 \le p \le m) \\ & \boldsymbol{X}_i \succeq O \qquad\qquad\qquad (1 \le i \le \ell) \end{array} \right. \\
\texttt{Dual} & \left\{ \begin{array}{ll} \textbf{maximize} & \sum_{p=1}^{m} b_p y_p \\ \textbf{subject to} & \sum_{p=1}^{m} \boldsymbol{A}_{ip} y_p + \boldsymbol{S}_i = \boldsymbol{C}_i \quad (1 \le i \le \ell) \\ & \boldsymbol{S}_i \succeq O \qquad\qquad\qquad (1 \le i \le \ell) \end{array} \right.
\end{array}
$$



- where $\boldsymbol{C}_i, \boldsymbol{A}_{i1}, \cdots, \boldsymbol{A}_{im} \in \mathcal{S}^{n_i}$ $(1 \le i \le \ell)$, $\boldsymbol{b} \in \mathbb{R}^m$ are given
- admits multiple block matrices

$\mathcal{S}^n$	:	space of $n \times n$-symmetric matrices
$\boldsymbol{X}_i \in \mathcal{S}^{n_i}$	:	primal matrix variables
$\boldsymbol{S}_i \in \mathcal{S}^{n_i}$	:	dual matrix variables, $\boldsymbol{y} \in \mathbb{R}^m$: dual vector variable
$\boldsymbol{X} \succeq O$	:	$\boldsymbol{X}$ is symmetric positive semidefinite matrix

Existing Methods and General Solvers for SDPs

(I) Primal-dual path-following interior-point methods

- general formulation, several search directions: NT, H..K..M, *etc.*
- CSDP *6.0.1*, SDPA *7.1.0*, SDPT3 *4.0β*, SeDuMi *1.1R3*, SDPARA *1.0.1*

(Ia) Krylov Iterative Methods

- *(Nakata-Fujisawa-Kojima PISM'98, Lin-Saigal BIT'00, Toh-Kojima SIOPT '02, Toh SIOPT '03)*

(II) Dual interior-point methods

- uses only dual variables
- DSDP *5.8* *(S.Benson-Ye-Y.Zhang SIOPT'00)*

(III) Spectral Bundle method

- SBmethod *1.1.3* *(Helmberg-Rendl SIOPT'00)*

(IV) Nonlinear formulation

- PENNON *(Kočvara-Stingl OMS'02)*
- SDPLR *1.02* (*Burer-Monteiro MPb'03)*
- etc.

Interior-Point Methods for SDPs

$\mu > 0$
$(\mu \to 0)$

$$\text{Primal} \left\{ \begin{array}{lll} \textbf{minimize} & \sum_{i=1}^{\ell} \text{tr}(\boldsymbol{C}_i \boldsymbol{X}_i) - \mu \log\det \boldsymbol{X}_i & \\ \textbf{subject to} & \sum_{i=1}^{\ell} \text{tr}(\boldsymbol{A}_{ip} \boldsymbol{X}_i) = b_p & (1 \le p \le m) \\ & \boldsymbol{X}_i \succ O & (1 \le i \le \ell) \end{array} \right.$$

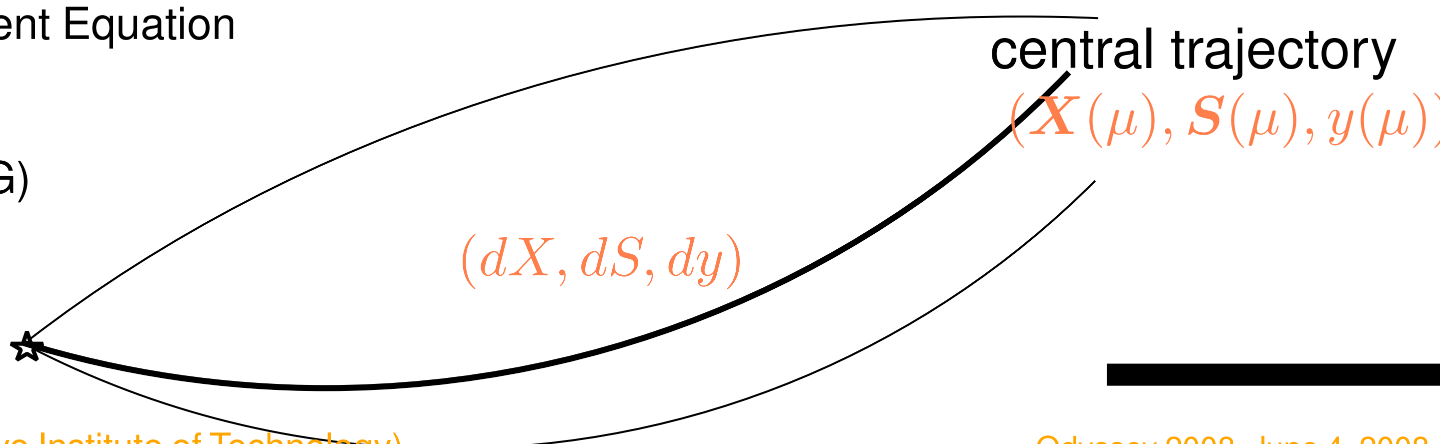
$$\text{Dual} \left\{ \begin{array}{lll} \textbf{maximize} & \sum_{p=1}^{m} b_p y_p + \sum_{i=1}^{\ell} \mu \log\det \boldsymbol{S}_i \quad \text{+constant} & \\ \textbf{subject to} & \sum_{p=1}^{m} \boldsymbol{A}_{ip} y_p + \boldsymbol{S}_i = \boldsymbol{C}_i & (1 \le i \le \ell) \\ & \boldsymbol{S}_i \succ O & (1 \le i \le \ell) \end{array} \right.$$

Optimality conditions

$$\begin{array}{lll} \boldsymbol{X}_i \succ O, & \sum_{i=1}^{\ell} \text{tr}(\boldsymbol{A}_{ip} \boldsymbol{X}_i) = b_p & (1 \le p \le m) \\ \boldsymbol{S}_i \succ O, & \sum_{p=1}^{m} \boldsymbol{A}_{ip} y_p + \boldsymbol{S}_i = \boldsymbol{C}_i & (1 \le i \le \ell) \\ \boldsymbol{X}_i \boldsymbol{S}_i = \mu \boldsymbol{I} & & \end{array}$$

Schur Complement Equation

$$\left\{ \begin{array}{l} \boldsymbol{M} dy = \boldsymbol{u} \\ \text{(Cholesky/CG)} \end{array} \right.$$



$(\mu \longrightarrow 0)$
optimal solution

Interior-Point Methods for SDPs

$\mu > 0$
$(\mu \to 0)$

$$
\texttt{Primal} \begin{cases} \textbf{minimize} & \sum_{i=1}^{\ell} \mathrm{tr}(\boldsymbol{C}_i \boldsymbol{X}_i) - \mu \log\det \boldsymbol{X}_i \\ \textbf{subject to} & \sum_{i=1}^{\ell} \mathrm{tr}(\boldsymbol{A}_{ip} \boldsymbol{X}_i) = b_p \quad (1 \le p \le m) \\ & \boldsymbol{X}_i \succ O \quad (1 \le i \le \ell) \end{cases}
$$

$$
\texttt{Dual} \begin{cases} \textbf{maximize} & \sum_{p=1}^{m} b_p y_p + \sum_{i=1}^{\ell} \mu \log\det \boldsymbol{S}_i \quad +\text{constant} \\ \textbf{subject to} & \sum_{p=1}^{m} \boldsymbol{A}_{ip} y_p + \boldsymbol{S}_i = \boldsymbol{C}_i \quad (1 \le i \le \ell) \\ & \boldsymbol{S}_i \succ O \quad (1 \le i \le \ell) \end{cases}
$$

Optimality conditions

$$
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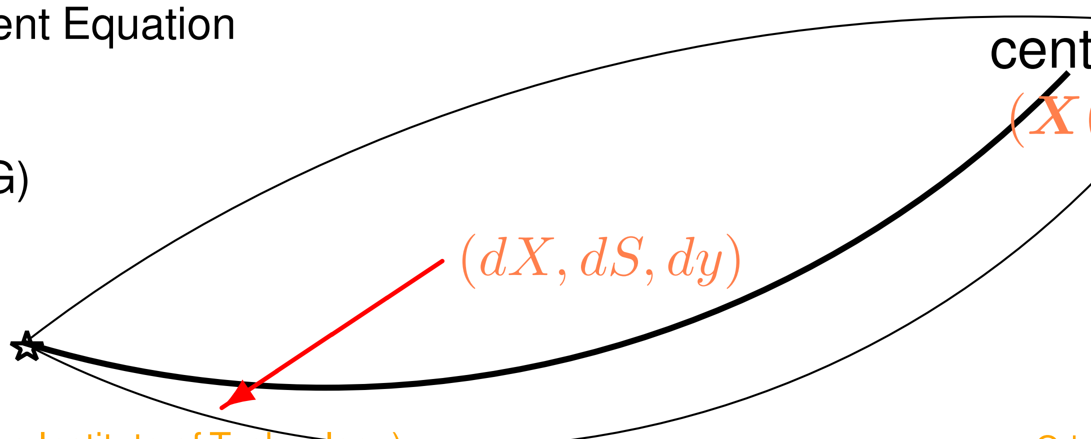

Interior-Point Methods for SDPs

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$(\mu \to 0)$

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\end{array}\right.
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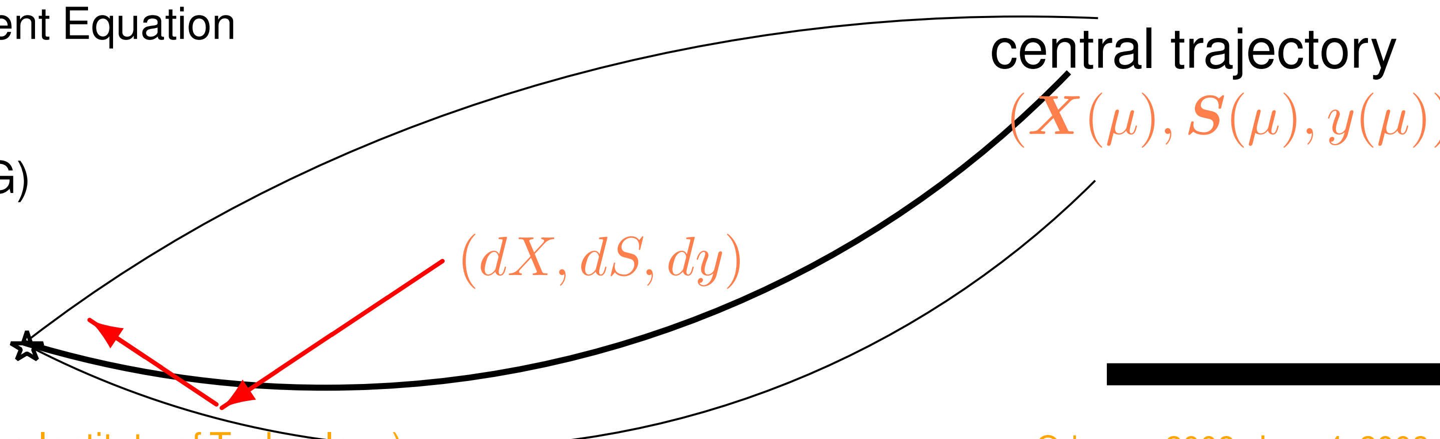

Interior-Point Methods for SDPs

$\mu > 0$
$(\mu \to 0)$

$$
\text{Primal} \begin{cases} \textbf{minimize} & \sum_{i=1}^{\ell} \mathrm{tr}(\boldsymbol{C}_i \boldsymbol{X}_i) - \mu \log\det \boldsymbol{X}_i & \\ \textbf{subject to} & \sum_{i=1}^{\ell} \mathrm{tr}(\boldsymbol{A}_{ip} \boldsymbol{X}_i) = b_p & (1 \le p \le m) \\ & \boldsymbol{X}_i \succ O & (1 \le i \le \ell) \end{cases}
$$

$$
\text{Dual} \begin{cases} \textbf{maximize} & \sum_{p=1}^{m} b_p y_p + \sum_{i=1}^{\ell} \mu \log\det \boldsymbol{S}_i \quad +\text{constant} & \\ \textbf{subject to} & \sum_{p=1}^{m} \boldsymbol{A}_{ip} y_p + \boldsymbol{S}_i = \boldsymbol{C}_i & (1 \le i \le \ell) \\ & \boldsymbol{S}_i \succ O & (1 \le i \le \ell) \end{cases}
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Optimality conditions

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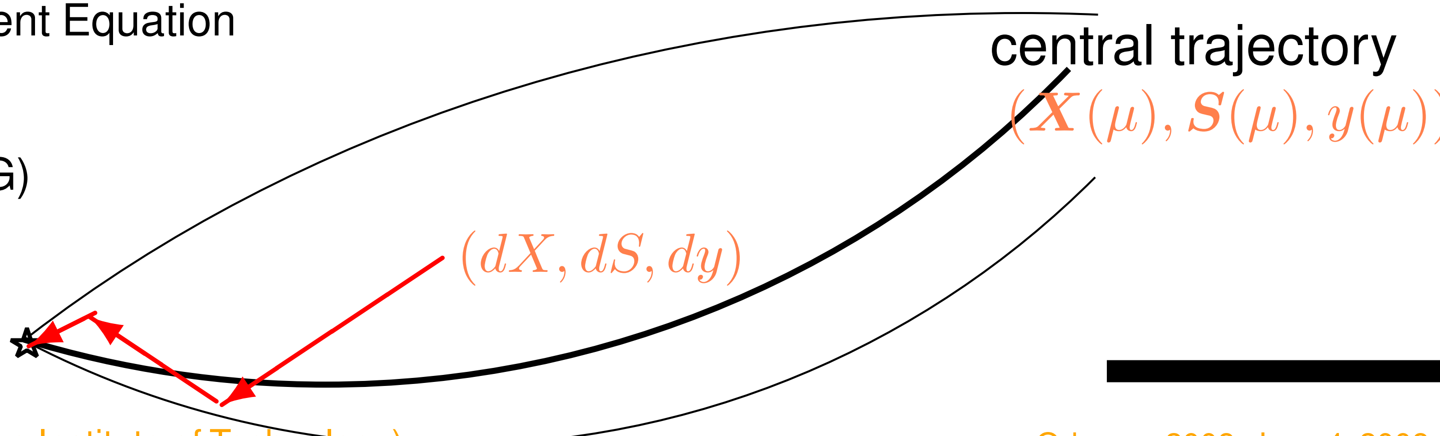


$(\mu \longrightarrow 0)$
optimal solution

Primal and Dual SDP Formulations

- SDP software only accepts problems in a specific formulation

$\Uparrow$ critical restriction

$$
\text{Primal} \left\{ \begin{array}{lll} \textbf{minimize} & \sum_{i=1}^{\ell} \mathrm{tr}(\boldsymbol{C}_i \boldsymbol{X}_i) & \\ \textbf{subject to} & \sum_{i=1}^{\ell} \mathrm{tr}(\boldsymbol{A}_{ip} \boldsymbol{X}_i) = b_p & (1 \le p \le m) \\ & \boldsymbol{X}_i \succeq O & (1 \le i \le \ell) \end{array} \right.
$$

$$
\text{Dual} \left\{ \begin{array}{lll} \textbf{maximize} & \sum_{p=1}^{m} b_p y_p & \\ \textbf{subject to} & \sum_{p=1}^{m} \boldsymbol{A}_{ip} y_p + \boldsymbol{S}_i = \boldsymbol{C}_i & (1 \le i \le \ell) \\ & \boldsymbol{S}_i \succeq O & (1 \le i \le \ell) \end{array} \right.
$$

- We can choose between formulate as a primal or dual SDP
- Examples of primal SDP formulation (and dual SDP formulation)

Fermionic System with N electrons with 1-RDM (1/3)

$$\begin{cases} \text{minimize} & \mathrm{tr}(H_1\gamma) \\ \text{subject to} & \mathrm{tr}(\hat{N}\gamma) = N \\ & \gamma \succeq \mathbf{0} \\ & \boldsymbol{I} - \gamma \succeq \mathbf{0} \end{cases}$$

where

r	:	spin orbitals or rank
$\gamma \in \mathcal{S}^r$	:	1-RDM
$H_1 \in \mathcal{S}^r$	:	one-body Hamiltonian
$\hat{N}$	:	number operator
$\succeq \mathbf{0}$	:	rhs matrix is positive semidefinite
$\boldsymbol{I}$	:	identity matrix

Fermionic System with N electrons with 1-RDM (2/3)

$$\left\{\begin{array}{l}\gamma \succeq \mathbf{0} \\ \boldsymbol{I}-\gamma \succeq \mathbf{0}\end{array}\right. \Leftrightarrow \tilde{\gamma} = \begin{pmatrix} \tilde{\gamma}_1 & 0 \\ 0 & \tilde{\gamma}_2 \end{pmatrix} = \begin{pmatrix} \gamma & 0 \\ 0 & \boldsymbol{I}-\gamma \end{pmatrix} \succeq \mathbf{0}$$

$$\Rightarrow [\tilde{\gamma}_1]_{ij} + [\tilde{\gamma}_2]_{ij} = \delta^i_j, \qquad i,j = 1,2,\ldots,r$$

$$\tilde{H}_1 = \begin{pmatrix} H_1 & 0 \\ 0 & 0 \end{pmatrix}, \quad \tilde{N} = \begin{pmatrix} \hat{N} & 0 \\ 0 & 0 \end{pmatrix}, \quad \boldsymbol{A}_{ij} = \begin{pmatrix} \boldsymbol{E}_{ij} & 0 \\ 0 & \boldsymbol{E}_{ij} \end{pmatrix}$$

where

$$\boldsymbol{E}_{ij} = \begin{cases} 1, & \text{for } (i,i) \\ 1/2, & \text{for } (i,j) \text{ or } (j,i),\ i<j \end{cases}$$

$$\begin{aligned} &\Rightarrow \quad \mathrm{tr}(\tilde{H}_1\tilde{\gamma}) = \mathrm{tr}(H_1\gamma) \\ &\Rightarrow \quad \mathrm{tr}(\tilde{N}\tilde{\gamma}) = \mathrm{tr}(\hat{N}\gamma) = N \\ &\Rightarrow \quad \mathrm{tr}(\boldsymbol{A}_{ij}\tilde{\gamma}) = [\tilde{\gamma}_1]_{ij} + [\tilde{\gamma}_2]_{ij} = \delta^i_j \end{aligned}$$

Fermionic System with N electrons with 1-RDM (3/3)

Primal SDP Formulation

$$\begin{cases} \text{minimize} & \mathrm{tr}(\tilde{H}_1 \tilde{\gamma}) \\ \text{subject to} & \mathrm{tr}(\tilde{N}\tilde{\gamma}) = N \\ & \mathrm{tr}(\boldsymbol{A}_{ij}\tilde{\gamma}) = \delta^i_j, \quad 1 \leq i \leq j \leq r \\ & \tilde{\gamma} \succeq \mathbf{0} \end{cases}$$

$$\texttt{Primal} \begin{cases} \textbf{minimize} & \displaystyle\sum_{i=1}^{\ell} \mathrm{tr}(\boldsymbol{C}_i \boldsymbol{X}_i) \\ \textbf{subject to} & \displaystyle\sum_{i=1}^{\ell} \mathrm{tr}(\boldsymbol{A}_{ip} \boldsymbol{X}_i) = b_p \quad (1 \leq p \leq m) \\ & \boldsymbol{X}_i \succeq O \quad (1 \leq i \leq \ell) \end{cases}$$

Fermionic System with N electrons with 2-RDM and P,Q (1/3)

$$\left\{\begin{array}{ll} \text{minimize} & \mathrm{tr}(\boldsymbol{H}\Gamma) \\ \text{subject to} & \mathrm{tr}(\hat{N}\Gamma) = N \\ & \Gamma \succeq \mathbf{0}, \quad \boldsymbol{Q} \succeq \mathbf{0} \end{array}\right.$$

r	:	spin orbitals or rank
$\Gamma \in \mathcal{S}^{r^2}$	:	2-RDM
$\boldsymbol{H} \in \mathcal{S}^{r^2}$	:	2-body Hamiltonian
P	:	$2\Gamma^{i_1 i_2}_{j_1 j_2}$
Q	:	$(\delta^{i_1}_{j_1}\delta^{i_2}_{j_2} - \delta^{i_1}_{j_2}\delta^{i_2}_{j_1}) - (\delta^{i_1}_{j_1}\gamma^{i_2}_{j_2} + \delta^{i_2}_{j_2}\gamma^{i_1}_{j_1}) + (\delta^{i_1}_{j_2}\gamma^{i_2}_{j_1} + \delta^{i_2}_{j_1}\gamma^{i_1}_{j_2}) + 2\Gamma^{i_1 i_2}_{j_1 j_2}$

- $\boldsymbol{P}$, $\boldsymbol{Q}$ matrices have 4 indices, and need to be mapped to a 2 indices matrix

Fermionic System with N electrons with 2-RDM and P,Q (2/3)

$$\left\{ \begin{array}{l} \boldsymbol{P} = 2\Gamma \succeq \mathbf{0} \\ \boldsymbol{Q} \succeq 0 \end{array} \right. \Leftrightarrow \tilde{\Gamma} = \begin{pmatrix} \Gamma & 0 \\ 0 & \boldsymbol{Q} \end{pmatrix} \succeq \mathbf{0}, \text{ let } \tilde{\boldsymbol{H}} = \begin{pmatrix} \boldsymbol{H} & 0 \\ 0 & 0 \end{pmatrix},$$

$$\tilde{N} = \begin{pmatrix} \hat{N} & 0 \\ 0 & 0 \end{pmatrix}, \quad \boldsymbol{A}_{i_1 i_2, j_1 j_2} = \begin{pmatrix} \tilde{\boldsymbol{E}}_{i_1 i_2, j_1 j_2} - \boldsymbol{E}_{i_1 i_2, j_1 j_2} & 0 \\ 0 & \boldsymbol{E}_{i_1 i_2, j_1 j_2} \end{pmatrix}$$

where

$$\boldsymbol{E}_{i_1 i_2, j_1 j_2} = \begin{cases} 1, & \text{for } (i_1 + (i_2 - 1)r, i_1 + (i_2 - 1)r) \\ 1/2, & \text{for } (i_1 + (i_2 - 1)r, j_1 + (j_2 - 1)r) \\ 1/2, & \text{for } (j_1 + (j_2 - 1)r, i_1 + (i_2 - 1)r), \ i_1 < j_1 \end{cases}$$

$$\tilde{\boldsymbol{E}}_{i_1 i_2, j_1 j_2} = \sum_{k=1}^{r} (\delta_{j_1}^{i_1} \boldsymbol{E}_{i_2,k,j_2,k} + \delta_{j_2}^{i_2} \boldsymbol{E}_{i_1 k, j_1 k} - \delta_{j_2}^{i_1} \boldsymbol{E}_{i_2 k, j_1 k} - \delta_{j_1}^{i_2} \boldsymbol{E}_{i_1 k, j_2 k})/(N-1)$$

Fermionic System with N electrons with 2-RDM and P,Q (3/3)

Primal SDP Formulation

$$\left\{\begin{array}{ll} \text{minimize} & \mathrm{tr}(\tilde{\boldsymbol{H}}\tilde{\Gamma}) \\ \text{subject to} & \mathrm{tr}(\tilde{N}\tilde{\Gamma}) = N \\ & \mathrm{tr}(\boldsymbol{A}_{i_1 i_2, j_1 j_2}\tilde{\Gamma}) = \delta^{i_1}_{j_1}\delta^{i_2}_{j_2} - \delta^{i_1}_{j_2}\delta^{i_2}_{j_1}, \quad 1 \le i_1 \le j_1 \le r \\ & \qquad\qquad\qquad\qquad\qquad\qquad\qquad\qquad\quad 1 \le i_2 \le j_2 \le r \\ & \tilde{\Gamma} \succeq \mathbf{0} \end{array}\right.$$

$$\texttt{Primal} \left\{\begin{array}{lll} \textbf{minimize} & \displaystyle\sum_{i=1}^{\ell} \mathrm{tr}(\boldsymbol{C}_i \boldsymbol{X}_i) & \\ \textbf{subject to} & \displaystyle\sum_{i=1}^{\ell} \mathrm{tr}(\boldsymbol{A}_{ip} \boldsymbol{X}_i) = b_p & (1 \le p \le m) \\ & \boldsymbol{X}_i \succeq O & (1 \le i \le \ell) \end{array}\right.$$

SDP Problem Sizes

$$
\texttt{Primal}
\begin{cases}
\textbf{minimize} & \displaystyle\sum_{i=1}^{\ell} \mathrm{tr}(\boldsymbol{C}_i \boldsymbol{X}_i) & \\
\textbf{subject to} & \displaystyle\sum_{i=1}^{\ell} \mathrm{tr}(\boldsymbol{A}_{ip} \boldsymbol{X}_i) = b_p & (1 \le p \le m) \\
& \boldsymbol{X}_i \succeq O & (1 \le i \le \ell)
\end{cases}
$$

$\mathcal{S}^{n_i}$	:	space of $n_i \times n_i$-symmetric matrices
$\boldsymbol{X}_i \in \mathcal{S}^{n_i}$	:	primal matrix variables

- Size of an SDP: # of constraints m
  dimension of matrices n_i
- Also depends on the sparsity of the matrices $\boldsymbol{C}_i$ and $\boldsymbol{A}_{ip}$

SDP Sizes in Primal SDP Formulation

	dimension of matrices n_i
P	$(r/2)^2 \times (r/2)^2$ (1 block), $\binom{r/2}{2} \times \binom{r/2}{2}$ (2 blocks)
Q	$(r/2)^2 \times (r/2)^2$ (1 block), $\binom{r/2}{2} \times \binom{r/2}{2}$ (2 blocks)
G	$2(r/2)^2 \times 2(r/2)^2$ (1 block), $(r/2)^2 \times (r/2)^2$ (2 blocks)
	# of constraints m
P, Q, G	$5 + 3\binom{r^2/4+1}{2} + 2\binom{r(r/2-1)/4+1}{2} + \binom{r^2/2+1}{2}$

where $\binom{a}{b} = \frac{a!}{b!(a-b)!}$, r spin orbitals or rank

SDP Sizes in Primal SDP Formulation

	P, Q, G		$P, Q, G, T1$		$P, Q, G, T1, T2'$	
r	m	n_i (max)	m	n_i (max)	m	n_i (max)
8	983	32	1603	24	10971	92
10	2365	50	5025	50	40685	180
12	4871	72	13481	90	120449	312
14	8993	98	32009	147	303385	497
16	15313	128	68905	224	677241	744
18	24503	162	136943	324	1377071	1062
20	37325	200	254795	450	2599915	1460
22	54631	242	448651	605	4621479	1947
24	77363	288	754039	792	7814815	2532
26	106553	338	1217845	1014	12671001	3224
28	143323	392	1900533	1274	19821821	4032
30	188885	450	2878565	1575	30064445	4965

Dual SDP Formulation

$$\begin{cases} \text{minimize} & \mathrm{tr}(H_1\gamma) + \mathrm{tr}(H_2\Gamma) \\ \text{subject to} & P, Q, G, T1, T2' \text{ conditions} \end{cases}$$

$\Downarrow$ γ, Γ corresponds to the variable $\boldsymbol{y}$

$$\boxed{\texttt{Dual} \quad \begin{cases} \textbf{maximize} & \sum_{p=1}^{m} b_p y_p & \\ \textbf{subject to} & \sum_{p=1}^{m} \boldsymbol{A}_{ip} y_p + \boldsymbol{S}_i = \boldsymbol{C}_i & (1 \le i \le \ell) \\ & \boldsymbol{S}_i \succeq O & (1 \le i \le \ell) \end{cases}}$$

SDP Sizes in Dual SDP Formulation

	dimension of matrices n_i
P	$(r/2)^2 \times (r/2)^2$ (1 block), $\binom{r/2}{2} \times \binom{r/2}{2}$ (2 blocks)
Q	$(r/2)^2 \times (r/2)^2$ (1 block), $\binom{r/2}{2} \times \binom{r/2}{2}$ (2 blocks)
G	$2(r/2)^2 \times 2(r/2)^2$ (1 block), $(r/2)^2 \times (r/2)^2$ (2 blocks)
$T1$	$\frac{r}{2}\binom{r/2}{2} \times \frac{r}{2}\binom{r/2}{2}$ (2 blocks), $\binom{r/2}{3} \times \binom{r/2}{3}$ (2 blocks)
	# of constraints m
any	$\binom{r^2/4+1}{2} + 2\binom{r(r/2-1)/4+1}{2}$ and 5

where $\binom{a}{b} = \frac{a!}{b!(a-b)!}$, $\quad r$ spin orbitals or rank

SDP Sizes in Dual SDP Formulation

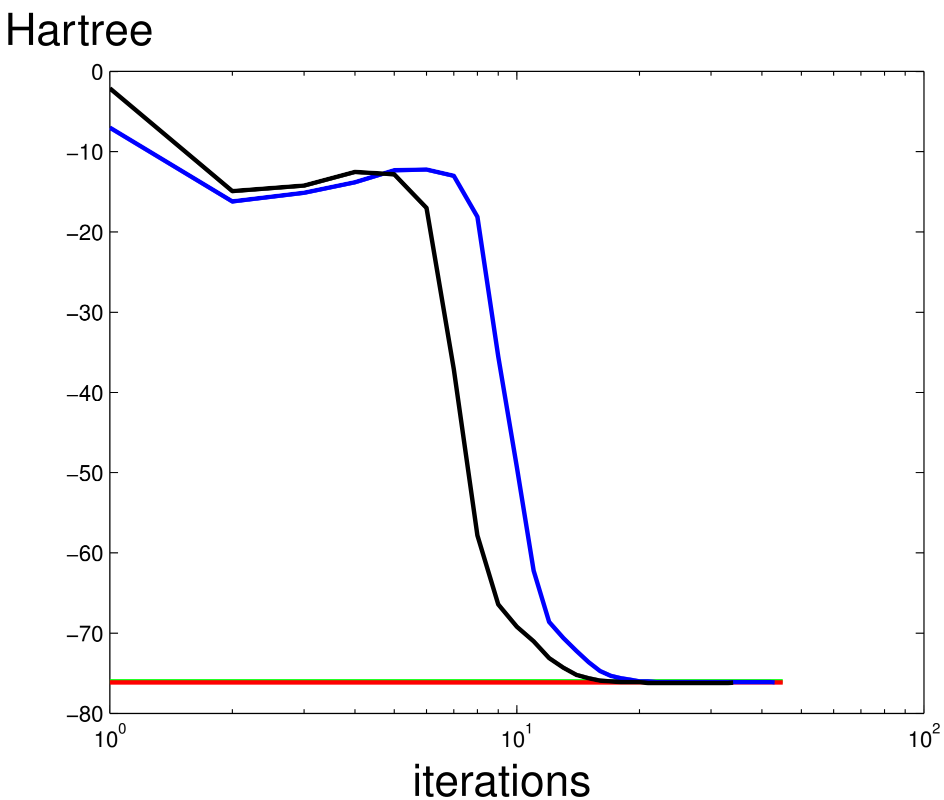
r	m	s	P,Q,G n_i (max)	$P,Q,G,T1$ n_i (max)	$P,Q,G,T1,T2'$ n_i (max)
8	178	5	32	24	92
10	435	5	50	50	180
12	906	5	72	90	312
14	1687	5	98	147	497
16	2892	5	128	224	744
18	4653	5	162	324	1062
20	7120	5	200	450	1460
22	10461	5	242	605	1947
24	14862	5	288	792	2532
26	20527	5	338	1014	3224
28	27678	5	392	1274	4032
30	36555	5	450	1575	4965

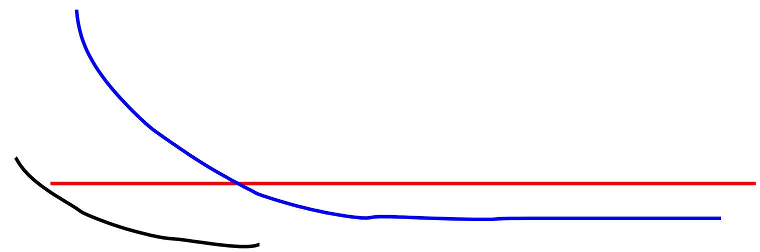
Primal SDP Formulation x Dual SDP Formulation

		$r = 10$		$r = 20$		$r = 30$	
		m	n_i (max)	m	n_i (max)	m	n_i (max)
P, Q, G	primal	2,365	50	37,325	200	188,885	450
	dual	435	50	7,120	200	36,555	450
P, Q, G	primal	5,025	50	254,795	450	2,878,565	1,575
$T1$	dual	435	50	7,120	450	36,555	1,575
P, Q, G	primal	40,685	180	2,599,915	1,460	30,064,445	4,965
$T1, T2'$	dual	435	180	7,120	1,460	36,555	4,965

- The number of constraints m on the dual SDP formulation does not depend on the N-representability conditions

Number of Iterations by Interior-Point Method for H_2O (double-ζ, $r = 28$) with PGQ,$PQGT1T2'$





- Interior-Point Methods always converge regardless of the chosen initial point
- Each iteration is very cost, but its convergence is extremely fast

RRSDP Method (D. A. Mazziotti)

- parallel interior-point methods seems the correct approach
  $\Rightarrow$ because need to solve SDPs with high accuracy ... BUT
- D. A. Mazziotti, "Realization of quantum chemistry without wave functions through first-order semidefinite programming", Physical Review Letters **(93)** 213001 (2004)

$\Longrightarrow$ first-order method (RRSDP)

$$\begin{cases} \text{minimize} & \sum_{i=1}^{\ell} \mathrm{tr}(\boldsymbol{C}\boldsymbol{X}_i) \\ \text{s.t.} & \sum_{i=1}^{\ell} \mathrm{tr}(\boldsymbol{A}_{ip}\boldsymbol{X}) = b_p \ (p = 1, \ldots, m), \ \boldsymbol{X}_i \succeq O \end{cases}$$

- $\mathcal{S}^{n_i} \ni \boldsymbol{X}_i = R_i R_i^T \succeq O$, where $R_i \in \mathbb{R}^{n_i \times n_i}$
- nonlinear problem $\Leftarrow$ augmented Lagrangian + L-BFGS
- very similar to Burer-Monteiro's low-rank factorization
- S. Burer, and R. D. C. Monteiro, "A nonlinear programming algorithm for solving semidefinite programs via low-rank factorization", Mathematical Programming **Series B 95** 329 (2003); S. Burer and R. D. C. Monteiro, "Local minima and convergence in low-rank semidefinite programming", Mathematical Programming **103** 427 (2005)

Floating-Point Operations and Memory Usage by PDIPM and RRSDP

N-representability conditions		P, Q, G		
formulation	algorithm	FLOPI	# iterations	memory
primal SDP formulation	PDIPM	r^{12}	$r \ln \varepsilon^{-1}$	r^8
	RRSDP	r^6	?	r^4
dual SDP formulation	PDIPM	r^{12}	$r \ln \varepsilon^{-1}$	r^8
	RRSDP	r^6	?	r^4

N-representability conditions		P, Q, G, $T1$ or P, Q, G, $T1$, $T2'$		
formulation	algorithm	FLOPI	# iterations	memory
primal SDP formulation	PDIPM	r^{18}	$r^{3/2} \ln \varepsilon^{-1}$	r^{12}
	RRSDP	r^9	?	r^6
dual SDP formulation	PDIPM	r^{12}	$r^{3/2} \ln \varepsilon^{-1}$	r^8
	RRSDP	r^9	?	r^6

Concluding Remarks

- From the limitations of the SDP software, the dual SDP formulation is the correct approach
- Very accurate values for the ground state energy for atoms and molecules and 1-D Hubbard Model can be calculated

  Today 4:15-4:45 Maho Nakata, “The Reduced Density Matrix Method: Applications of $T2'$ N-representability Conditions and Development of Highly Accurate Solver”
- There is a severe limit on the size of the systems due to the SDP problem size ($r = 28$ with P, Q, G, $T1$, $T2'$ conditions)