Perturbative calculation of Jastrow factor

Hongjun Luo and Wolfgang Hackbusch
Max-Planck-Institut für Mathematik in den Naturwissenschaften, Leipzig

Heinz-Jürgen Flad
Institut für Informatik, Christian-Albrechts-Universität zu Kiel, Kiel

Dietmar Kolb
Fachbereich Naturwissenschaften der Universität Kassel

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Jastrow ansatz for electron correlation

Size consistent exponential ansatz:

$$\Psi(\mathbf{x_1}, \cdots, \mathbf{x_N}) = e^{\tau(\mathbf{x_1}, \cdots, \mathbf{x_N})} \Phi(\mathbf{x_1}, \cdots, \mathbf{x_N})$$

Φ: Mean field solution (Hartree Fock)

- Compact description of electron correlation
- Intensively used in quantum Monte Carlo calculations
- Can be treated by multiscale methods (wavelet, sparse grid)

Available methods

- FHNC: difficult to implement
- VMC optimization: not efficient for complex trial functions

Alternative approach: Systematic perturbation theory.

Perturbation theory for Jastrow factor

Similar to coupled-cluster perturbation theory

T. Helgaker, P. Jørgensen and J. Olsen, *Molecular Electronic-Structure Theory*, (Wiley, New York 1999)

Variational minimization of the energy in the space of Jastrow functions

$$\Psi(\mathbf{R}) = F(\mathbf{R})\Phi(\mathbf{R}) = e^{\tau(\mathbf{R})}\Phi, \quad \mathbf{R} = (\mathbf{x}_1, \mathbf{x}_2, \cdots, \mathbf{x}_N),$$

$$\mathbf{E} = \inf_{\tau} \frac{< e^{\tau^\dagger} \hat{H} e^{\tau} >}{< e^{\tau^\dagger} e^{\tau} >},$$

where $< \cdots > \equiv < \Phi | \cdots | \Phi >$.

Expansion of $\tau(\mathbf{R})$ in permutationally symmetric bases

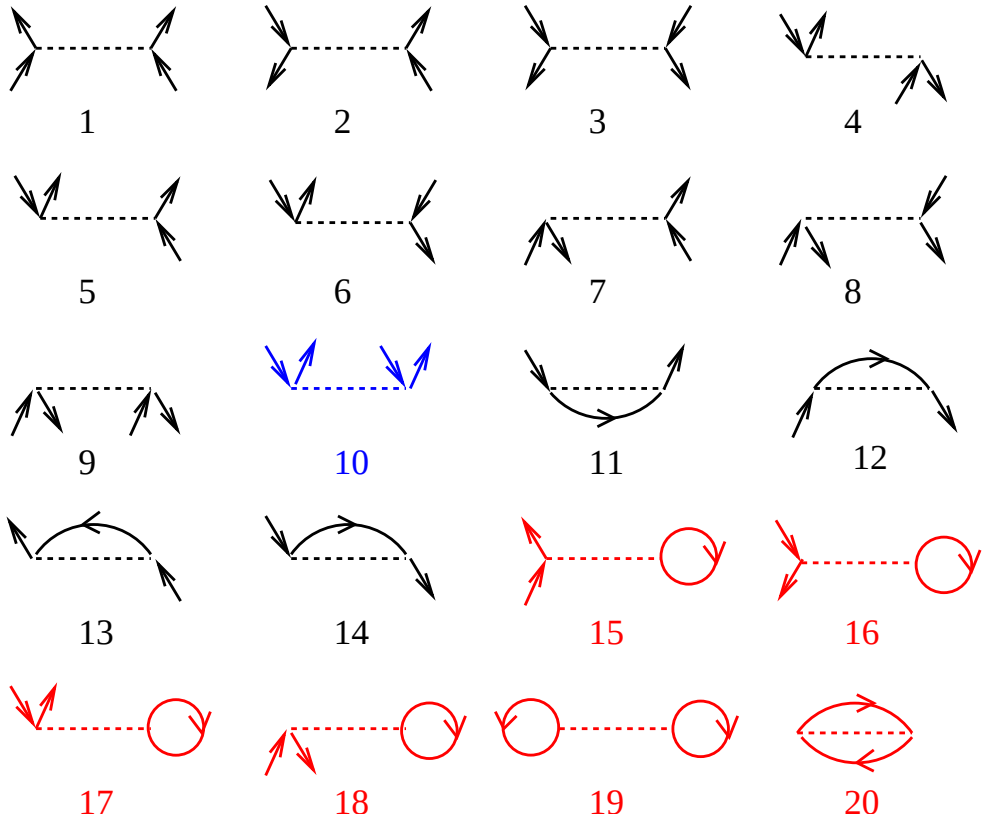
$$\hat{\tau}(\mathbf{R}) = \sum_m \sum_\alpha a_\alpha \hat{U}_\alpha^{(m)}(\mathbf{R})$$

$$U_0^{(0)}(\mathbf{R}) = 1, \quad U_\alpha^{(1)}(\mathbf{R}) = \sum_i u_\alpha^{(1)}(\mathbf{x}_i), \quad U_\beta^{(2)}(\mathbf{R}) = \frac{1}{2} \sum_{i \neq j} u_\beta^{(2)}(\mathbf{x}_i, \mathbf{x}_j), \cdots$$

Represent $U_\alpha^{(m)}$ in second quantisation and neglect diagrams which cause linear dependences

Follwing the idea of G. Stollhoff and P. Fulde, J. Chem. Phys. 73 (1980) 4548.

$$U_\alpha^{(2)} = \frac{1}{2}\sum_{i\neq j} u_\alpha^{(2)}(\mathbf{x}_i, \mathbf{x}_j) = \frac{1}{2}\sum_{pqrs} < pq|u_\alpha^{(2)}|rs > c_p^\dagger c_q^\dagger c_s c_r,$$



Diagrammatic representation of all possible terms in $\hat{U}_\alpha^{(2)}$.

non-linear eigenvalue problem

$$< \hat{U}_\alpha^{(m)\dagger} e^{\tau^\dagger} \hat{H} e^{\tau} >= E < \hat{U}_\alpha^{(m)\dagger} e^{\tau^\dagger} e^{\tau} >$$

Perturbation treatment

$$\hat{H}^\lambda = \hat{H}_0 + \lambda \hat{W},$$

$$\begin{aligned} E(\lambda) &= E_0 + E_1\lambda + E_2\lambda^2 + \cdots + E_K\lambda^K + \cdots, \\ \hat{\tau}(\lambda) &= \hat{\tau}_0 + \hat{\tau}_1\lambda + \hat{\tau}_2\lambda^2 + \cdots + \hat{\tau}_K\lambda^K + \cdots. \end{aligned}$$

expansion of the exponential function

$$\begin{aligned} e^{\hat{\tau}(\lambda)} &= \hat{F}_0 + \hat{F}_1\lambda + \hat{F}_2\lambda^2 + \cdots + \hat{F}_K\lambda^K + \cdots, \\ \hat{F}_0 &= 1, \quad (\hat{\tau}_0 = 0), \\ \hat{F}_1 &= \hat{\tau}_1, \\ \hat{F}_2 &= \hat{\tau}_2 + \frac{1}{2}\hat{\tau}_1^2 \ , \\ \hat{F}_3 &= \hat{\tau}_3 + \hat{\tau}_2 \ \hat{\tau}_1 + \frac{1}{3!}\hat{\tau}_1^3, \\ \hat{F}_4 &= \hat{\tau}_4 + \hat{\tau}_3 \ \hat{\tau}_1 + \frac{1}{2}\hat{\tau}_2^2 + \frac{1}{2}\hat{\tau}_2\hat{\tau}_1^2 + \frac{1}{4!}\hat{\tau}_1^4, \\ &\vdots \quad . \end{aligned}$$

$\hat{\tau}_1$ is only a pair correlation function, which does not refer to specific orbitals.

First order Jastrow factor

$$< \hat{U}_\alpha^{(m)\dagger}(\hat{H}_0 - E_0)\ \hat{\tau}_1 >= - < \hat{U}_\alpha^{(m)\dagger}\hat{W} >$$

Second and third order energies

$$\begin{aligned} E_2 &= < \hat{W}\hat{\tau}_1 >, \\ E_3 &= < \hat{\tau}_1^\dagger(\hat{W} - E_1)\hat{\tau}_1 > . \end{aligned}$$

Wigner's rule holds true.

Second order Jastrow factor

$$< \hat{U}_\alpha^{(m)\dagger}(\hat{H}_0 - E_0)\hat{\tau}_2 >= - < \hat{U}_\alpha^{(m)\dagger}(\hat{W} - E_1)\hat{\tau}_1 > -\frac{1}{2} < \hat{U}_\alpha^{(m)\dagger}(\hat{H}_0 - E_0)\hat{\tau}_1^2 >$$

Fouth order energy

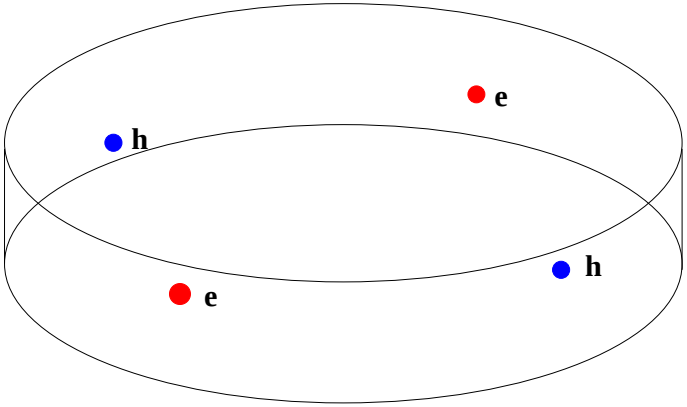
$$\begin{aligned} E_4 &= < (\hat{\tau}_2 + \frac{1}{2}\hat{\tau}_1^2)^\dagger(\hat{H}_0 - E_0)(\hat{\tau}_2 + \frac{1}{2}\hat{\tau}_1^2) > \\ &\quad +2 < \hat{\tau}_1^\dagger(\hat{W} - E_1)(\hat{\tau}_2 + \frac{1}{2}\hat{\tau}_1^2) > -E_2 < \hat{\tau}_1^\dagger\hat{\tau}_1 > . \end{aligned}$$

Intended applications

Quasi-two dimensional (bi)excitons

excitons: bound states of electron-hole pairs in semiconductor hetero-structures, analogous to 2D hydrogen atom.

bi-excitons: bound states of two excitons, analogous to hydrogen molecule.



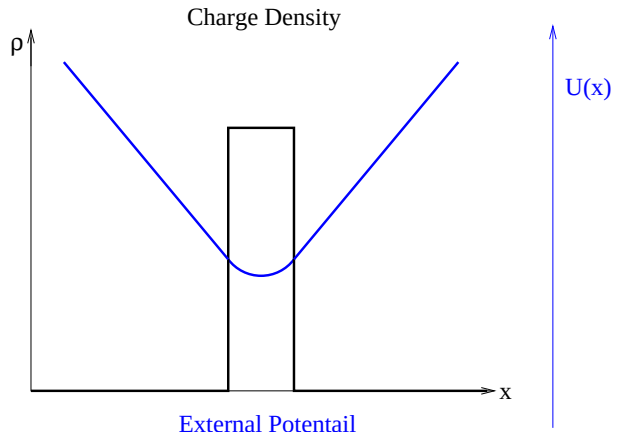
$$\begin{aligned}\hat{H} &= -\frac{\hbar^2}{2m_e^*}\sum_{i_e}\Delta_{i_e} - \frac{\hbar^2}{2m_h^*}\sum_{i_h}\Delta_{i_h} + \sum_{i_e} v_e(\mathbf{r}_{i_e}) + \sum_{i_h} v_h(\mathbf{r}_{i_h}) \\ &\quad -\sum_{i_e,i_h}\frac{e^2}{\epsilon|\mathbf{r}_{i_e}-\mathbf{r}_{i_h}|} + \frac{1}{2}\sum_{i_e,j_e}\frac{e^2}{\epsilon|\mathbf{r}_{i_e}-\mathbf{r}_{j_e}|} + \frac{1}{2}\sum_{i_h,j_h}\frac{e^2}{\epsilon|\mathbf{r}_{i_h}-\mathbf{r}_{j_h}|}\end{aligned}$$

Special feature: m_e^* and m_h^* comparable, Born-Oppenheimer approxima-tion not applicable. FCI calculation converges slowly.

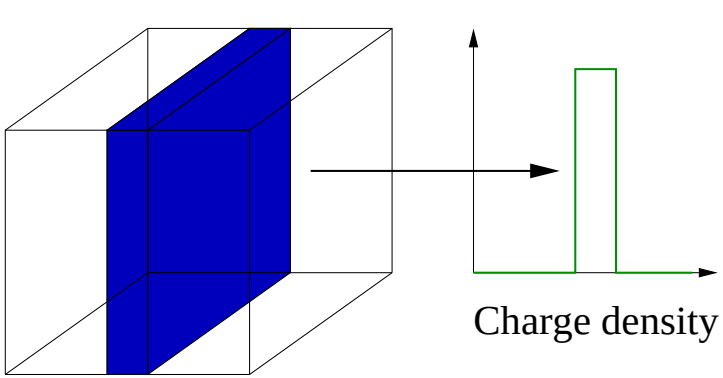
J. Shumway and D. M. Ceperley, Solid State Comm. 134 (2005)19

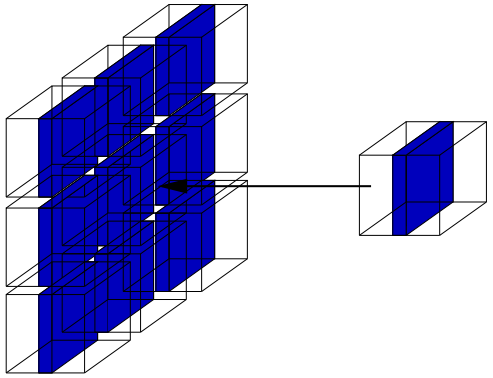
Intended: JPT calculations, VMC calculations with JPT Jastrow factor.

Jellium slab model



Supercell approach





$$\hat{H} = -\frac{1}{2}\sum_{i=1}^{N}\Delta_i + \sum_{i=1}^{N}U(\mathbf{r}_i) + \sum_{\mathbf{R}_s}\sum_{i<j}^{N}\frac{1}{|\mathbf{r}_i - \mathbf{r}_j - \mathbf{R}_s|}$$

$\mathbf{R}_s$: translation vectors of supercell lattice

RPA for inhomogeneuos systems

Gaudoin, Nekovee and Foukes, PRB 63(2001)115115

RPA of Bohm and Pines [PR 92 (1953) 609] can be generalized to inhomogeneuos systems, which leads to two decoupled Hamiltonians:

Short ranged electron Hamiltonian

$$\hat{H}_{sr} = \frac{1}{2}\sum \hat{\mathbf{p}}_i^2 + 2\pi \sum_{k>k_c} \frac{\hat{n}_{\mathbf{k}}\hat{n}_{\mathbf{k}}^\dagger}{k^2} - \frac{2\pi N}{V}\sum_{\mathbf{k}} \frac{1}{k^2} + \sum_i \tilde{V}(\hat{\mathbf{r}}_i),$$

Plasmon Hamiltonian

$$\hat{H}_p = \frac{1}{2}(\hat{\pi}\cdot\hat{\pi}^\dagger + \hat{\mathbf{q}}\cdot M\cdot\hat{\mathbf{q}}^\dagger).$$

JPT calculations can be applied to $\hat{H}_{sr}$.

Test calculation on screened Fermi gas

Periodic model of screened Fermi gas

$$\hat{H} = -\frac{1}{2}\sum_{i}^{N} \nabla_i^2 + \frac{1}{2}\sum_{\mathbf{R}} \sum_{i \neq j} v(\mathbf{r}_{ij} + \mathbf{R}), \qquad v(\mathbf{r}_{ij}) = \frac{e^{-\mu|\mathbf{r}_{ij}|}}{|\mathbf{r}_{ij}|}$$

Even tempered Gaussian bases

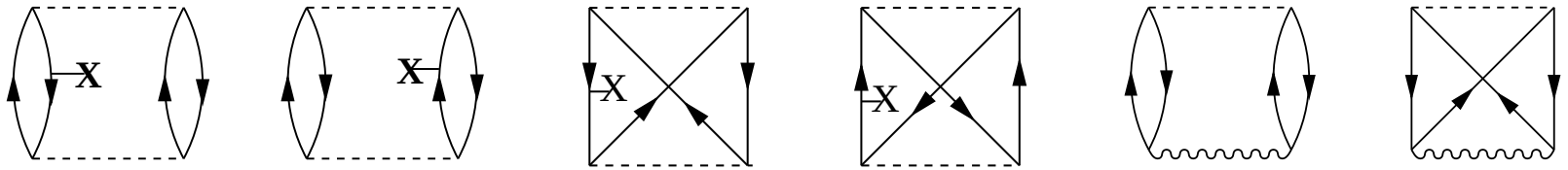
$$\hat{U}_\alpha(\mathbf{R}) = \frac{1}{2}\sum_{ij}^{N} u_\alpha^{(2)}(\mathbf{x}_i, \mathbf{x}_j), \qquad u_\alpha^{(2)}(\mathbf{x}, \mathbf{y}) = \sum_{\mathbf{R}} \exp(-\xi_\alpha(\mathbf{x} - \mathbf{y} + \mathbf{R})^2).$$

Perturbation calculation of the first order Jastrow factor $\hat{\tau}_1 = \sum_\alpha a_\alpha \hat{U}_\alpha$

Similar to Talman's linked cluster expansion on electron gas (PRA 10 (1974)1333)

$$\sum_\beta H^0_{\alpha\beta} a_\beta = -W_\alpha, \qquad H^0_{\alpha\beta} = < \hat{U}_\alpha{}^\dagger (\hat{H}^0 - E_0) \hat{U}_\beta >, \qquad W_\alpha = < \hat{U}_\alpha{}^\dagger \hat{W} >$$

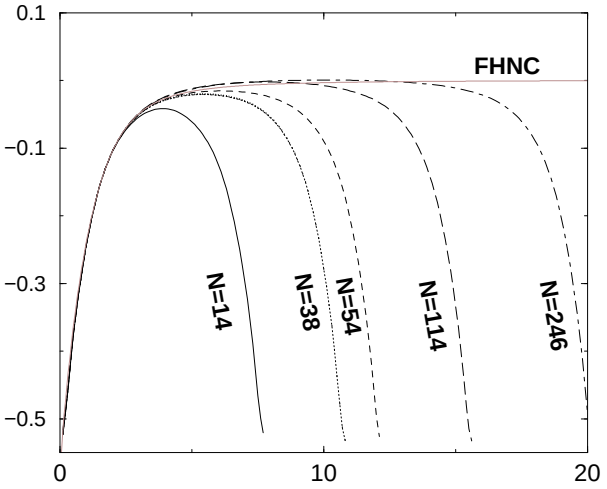
Diagrammatic calculation of the matrix elements



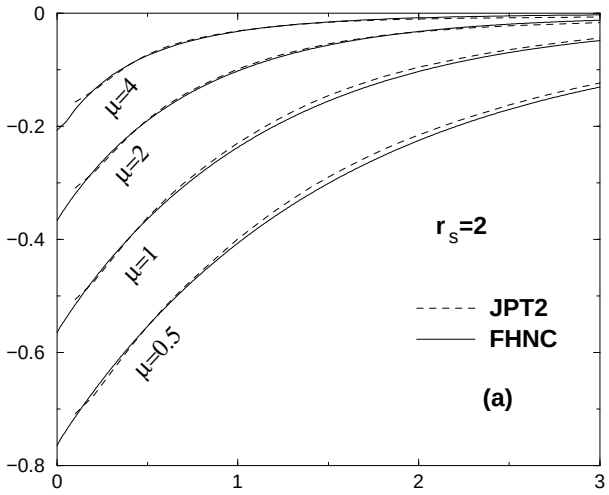
D1 **D2** D3 D4 **D5** **D6**

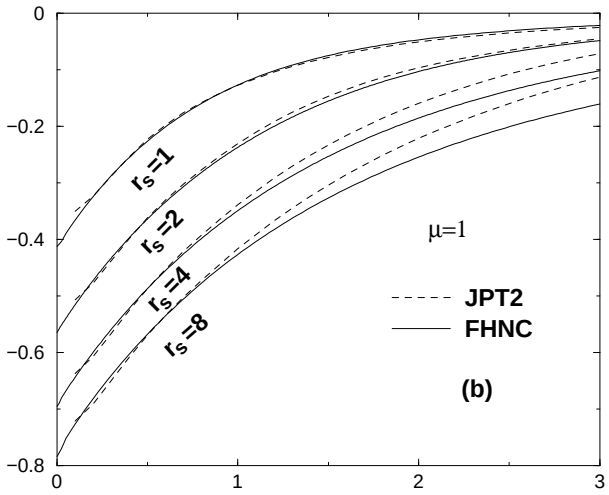
Compare first order Jastrow factor with FHNC results

Pair correlation Jastrow factor for different system sizes ($\mu = 1, r_s = 2$)

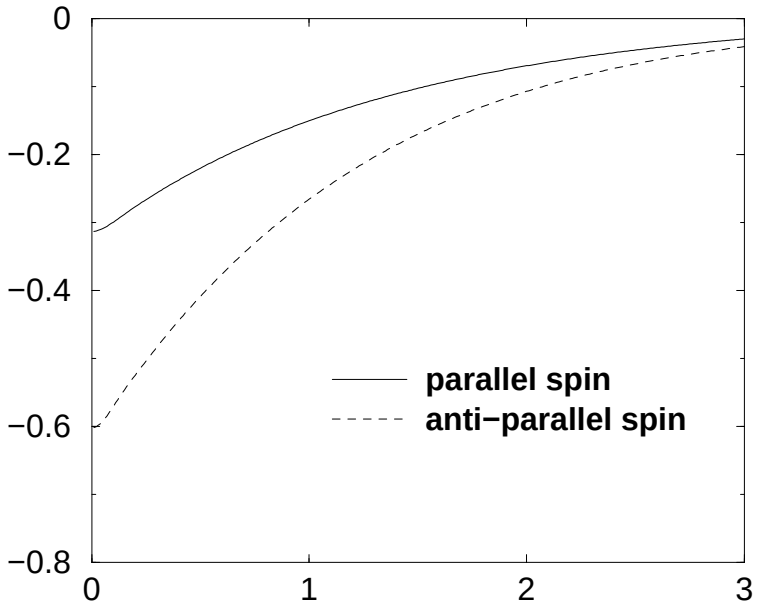


Jastrow factor for different screening and density ($N = 54$)





Spin dependent Jastrow factor ($\mu = 1, r_s = 2, N = 54$)



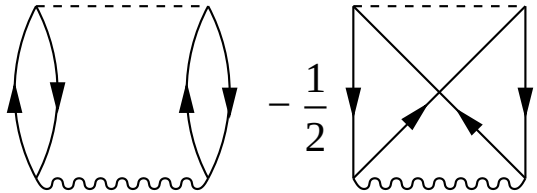
In good agreement with cusp condition

$$\left.\frac{dU}{dr}\right|_{r=0} = \begin{cases} 1/2 & \text{for opposite spins,} \\ 1/4 & \text{for same spins.} \end{cases}$$

Slopes estimated are 0.46 and 0.23 respectively.

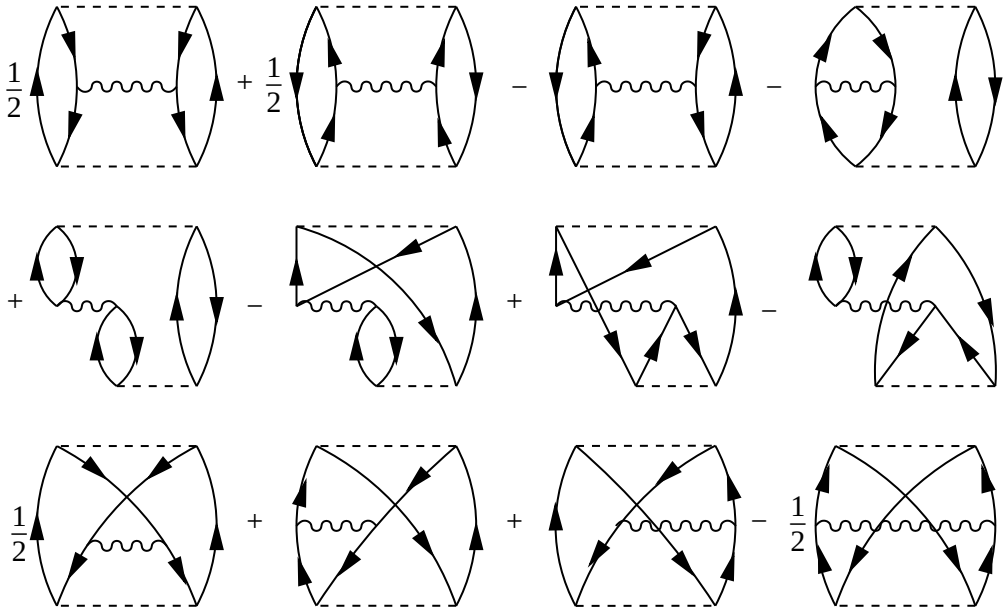
Second order perturbation energy

$$E_2 = < \hat{W}\hat{\tau}_1 >^c =$$



Third order perturbation energy (same complexity as Local ansatz)

$$E_3 = < \hat{\tau}_1^\dagger \hat{W}\hat{\tau}_1 >^c =$$



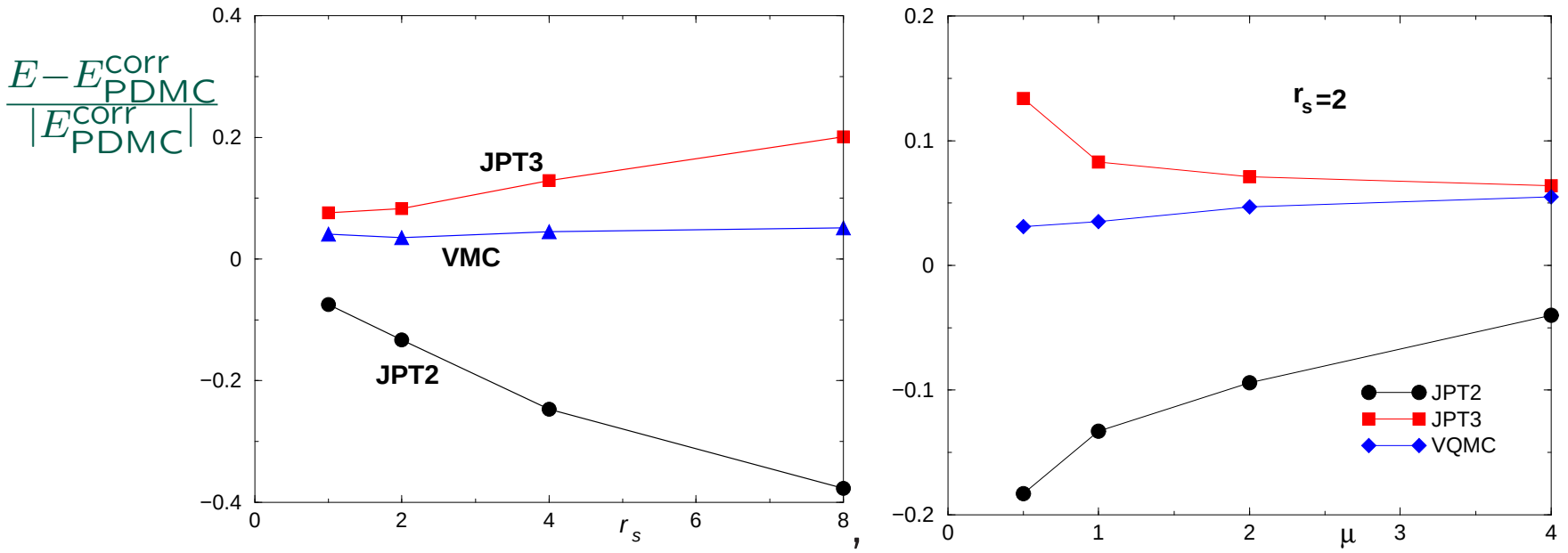
Benchmark calculation with pure diffusion Monte Carlo (PDMC) method

$$E^{\text{corr}}_{\text{PDMC}} \approx E^{\text{corr}}_{\text{exact}}$$

Variational Monte Carlo (VMC) evaluation of the Rayleigh-quotient

$$E^{\text{corr}}_{\text{VMC}} = \frac{< e^{\hat{\tau}_1^\dagger} \hat{H} e^{\hat{\tau}_1} >}{< e^{\hat{\tau}_1^\dagger} e^{\hat{\tau}_1} >} - E_{\text{HF}}$$

Compare the relative error of the correlation energy for different densities and different screening parameters



Conclusion

We have developed a perturbation method for the calculation of the correlation factor for the exponential Jastrow ansatz. Test calculations are performed for homogeneous Fermi gas models with screened Coulomb interaction. The results are judged by quantum Monte Carlo calculations (VMC, PDMC). Variation energy for the first order Jastrow factor can already produce 95% percent of the total correlation. The second and third order perturbation energies clearly converge to the variational ones.