

On the general mapping between effective two-electron interaction and Jastrow factors.

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This section presents the basics of the transcorrelation for electron-positron wave function

I. E-POSITRON JASTROW FACTOR

We assume that we have a Jastrow factor $j(\mathbf{r}_i, \mathbf{r}_p)$ where \mathbf{r}_i is the position of the i -th electron and \mathbf{r}_p is the position of the positron. We want to compute the similarity transformation of the following Hamiltonian

$$H_{ep} = -\frac{1}{2} \left(\Delta_{\mathbf{r}_p} + \sum_{i=1}^N (\Delta_{\mathbf{r}_i} + v_{ne}(\mathbf{r}_i)) \right) + \sum_{i>j} \frac{1}{r_{ij}} - \sum_i \frac{1}{r_{ip}}, \quad (1)$$

with the N -electron Jastrow factor

$$\begin{aligned} J(\mathbf{r}_1, \mathbf{r}_2, \dots, \mathbf{r}_N, \mathbf{r}_p) &= \sum_{i=1}^N j(\mathbf{r}_i, \mathbf{r}_p) \\ &= J(\{\mathbf{r}_i\}, \mathbf{r}_p) \end{aligned} \quad (2)$$

One can then use the usual Baker-Campbell-Hausdorff (BCH) expansion of the similarity transformation

$$e^{-J} H e^J = H + [H, J] + \frac{1}{2!} [[H, J], J] + \frac{1}{3!} [[[H, J], J], J] + \dots, \quad (3)$$

but because there is only a second order derivative operator in H_{ep} , it naturally terminates at second order

$$\begin{aligned} \tilde{H}_{ep}[J] &= e^{J(\{\mathbf{r}_i\}, \mathbf{r}_p)} H_{ep} e^{J(\{\mathbf{r}_i\}, \mathbf{r}_p)} \\ &= H_{ep} + [H_{ep}, J(\{\mathbf{r}_i\}, \mathbf{r}_p)] + \frac{1}{2} [[H_{ep}, J(\{\mathbf{r}_i\}, \mathbf{r}_p)], J(\{\mathbf{r}_i\}, \mathbf{r}_p)]. \end{aligned} \quad (4)$$

Actually, as $J(\{\mathbf{r}_i\}, \mathbf{r}_p)$ is a scalar function, the only part of H_{ep} that does not commute is the Laplacian. One can then write

$$\tilde{H}_{ep}[J] = H + [\hat{T}, J(\{\mathbf{r}_i\}, \mathbf{r}_p)] + \frac{1}{2} [[\hat{T}, J(\{\mathbf{r}_i\}, \mathbf{r}_p)], J(\{\mathbf{r}_i\}, \mathbf{r}_p)], \quad (5)$$

where \hat{T} is the total kinetic energy

$$\begin{aligned} \hat{T} &= -\frac{1}{2} (\Delta_{\mathbf{r}_p} + \sum_{j=1}^N \Delta_{\mathbf{r}_j}) \\ &= \hat{T}_p + \hat{T}_e. \end{aligned} \quad (6)$$

Let us compute the first-order commutator

$$[\hat{T}, J(\{\mathbf{r}_i\}, \mathbf{r}_p)] = [\hat{T}_p, J(\{\mathbf{r}_i\}, \mathbf{r}_p)] + [\hat{T}_e, J(\{\mathbf{r}_i\}, \mathbf{r}_p)]. \quad (7)$$

We begin by the electronic part by applying the commutator to a general electron-positron wave function $\phi(\{\mathbf{r}_i\}, \mathbf{r}_p)$

$$\begin{aligned} -2 \times [\hat{T}_e, J(\{\mathbf{r}_i\}, \mathbf{r}_p)] \phi(\{\mathbf{r}_i\}, \mathbf{r}_p) &= \\ \underbrace{\sum_{j=1}^N \Delta_{\mathbf{r}_j} \left(\sum_{i=1}^N j(\mathbf{r}_i, \mathbf{r}_p) \phi(\{\mathbf{r}_i\}, \mathbf{r}_p) \right)}_{=A} &- \underbrace{\sum_{i=1}^N j(\mathbf{r}_i, \mathbf{r}_p) \sum_{j=1}^N \Delta_{\mathbf{r}_j} \phi(\{\mathbf{r}_i\}, \mathbf{r}_p)}_{=B}. \end{aligned} \quad (8)$$

The term B can be split into three terms according to the x , y and z components of the Laplacian

$$B = B_x + B_y + B_z \quad (9)$$

where B_x is simply

$$B_x = \left(\sum_{i=1}^N j(\mathbf{r}_i, \mathbf{r}_p) \right) \left[\sum_{j=1}^N \frac{\partial^2}{\partial x_j^2} \phi(\{\mathbf{r}_i\}, \mathbf{r}_p) \right]. \quad (10)$$

Let us now proceed with the term A which can also be divided into three terms

$$A = A_x + A_y + A_z, \quad (11)$$

and let us take for example the A_x term

$$A_x = \sum_{j=1}^N \frac{\partial^2}{\partial x_j^2} \left(\sum_{i=1}^N j(\mathbf{r}_i, \mathbf{r}_p) \phi(\{\mathbf{r}_i\}, \mathbf{r}_p) \right). \quad (12)$$

and therefore the term $\sum_{i=1}^N$ in A_x reduces to $i = j$

$$\begin{aligned} A_x &= \sum_{j=1}^N \frac{\partial}{\partial x_j} \left(\left[\frac{\partial}{\partial x_j} \sum_{i=1}^N j(\mathbf{r}_i, \mathbf{r}_p) \right] \phi(\{\mathbf{r}_i\}, \mathbf{r}_p) \right. \\ &\quad \left. + \left(\sum_{i=1}^N j(\mathbf{r}_i, \mathbf{r}_p) \right) \left[\frac{\partial}{\partial x_j} \phi(\{\mathbf{r}_i\}, \mathbf{r}_p) \right] \right), \end{aligned} \quad (13)$$

but one can notice that

$$\frac{\partial}{\partial x_j} j(\mathbf{r}_i, \mathbf{r}_p) = 0 \quad \text{if } i \neq j, \quad (14)$$

and therefore

$$\left[\frac{\partial}{\partial x_j} \sum_{i=1}^N j(\mathbf{r}_i, \mathbf{r}_p) \right] \phi(\{\mathbf{r}_i\}, \mathbf{r}_p) = \left[\frac{\partial}{\partial x_j} j(\mathbf{r}_j, \mathbf{r}_p) \right] \phi(\{\mathbf{r}_i\}, \mathbf{r}_p) \quad (15)$$

and therefore

$$\begin{aligned} A_x &= \sum_{j=1}^N \frac{\partial}{\partial x_j} \left(\left[\frac{\partial}{\partial x_j} j(\mathbf{r}_j, \mathbf{r}_p) \right] \phi(\{\mathbf{r}_i\}, \mathbf{r}_p) \right. \\ &\quad \left. + \left(\sum_{i=1}^N j(\mathbf{r}_i, \mathbf{r}_p) \right) \left[\frac{\partial}{\partial x_j} \phi(\{\mathbf{r}_i\}, \mathbf{r}_p) \right] \right) \end{aligned} \quad (16)$$

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, or again

$$\begin{aligned}
A_x &= \sum_{j=1}^N \left(\left[\frac{\partial^2}{\partial x_j^2} j(\mathbf{r}_j, \mathbf{r}_p) \right] \phi(\{\mathbf{r}_i\}, \mathbf{r}_p) \right. \\
&\quad + \left[\frac{\partial}{\partial x_j} j(\mathbf{r}_j, \mathbf{r}_p) \right] \left[\frac{\partial}{\partial x_j} \phi(\{\mathbf{r}_i\}, \mathbf{r}_p) \right] \\
&\quad + \left[\frac{\partial}{\partial x_j} j(\mathbf{r}_j, \mathbf{r}_p) \right] \left[\frac{\partial}{\partial x_j} \phi(\{\mathbf{r}_i\}, \mathbf{r}_p) \right] \\
&\quad \left. + \left(\sum_{i=1}^N j(\mathbf{r}_i, \mathbf{r}_p) \right) \left[\frac{\partial^2}{\partial x_j^2} \phi(\{\mathbf{r}_i\}, \mathbf{r}_p) \right] \right) \\
&= B_x + \sum_{j=1}^N \left(\left[\frac{\partial^2}{\partial x_j^2} j(\mathbf{r}_j, \mathbf{r}_p) \right] \phi(\{\mathbf{r}_i\}, \mathbf{r}_p) \right. \\
&\quad \left. + 2 \left[\frac{\partial}{\partial x_j} j(\mathbf{r}_j, \mathbf{r}_p) \right] \left[\frac{\partial}{\partial x_j} \phi(\{\mathbf{r}_i\}, \mathbf{r}_p) \right] \right)
\end{aligned} \tag{17}$$

Therefore, by doing $A_x - B_x$, there only remains

$$\begin{aligned}
A_x - B_x &= \sum_{j=1}^N \left(\left[\frac{\partial^2}{\partial x_j^2} j(\mathbf{r}_j, \mathbf{r}_p) \right] \phi(\{\mathbf{r}_i\}, \mathbf{r}_p) \right. \\
&\quad \left. + 2 \left[\frac{\partial}{\partial x_j} j(\mathbf{r}_j, \mathbf{r}_p) \right] \left[\frac{\partial}{\partial x_j} \phi(\{\mathbf{r}_i\}, \mathbf{r}_p) \right] \right),
\end{aligned} \tag{18}$$

and we can then conclude that the commutator can be written as

$$\begin{aligned}
[\hat{T}_e, J(\{\mathbf{r}_i\}, \mathbf{r}_p)] \phi(\{\mathbf{r}_i\}, \mathbf{r}_p) &= \\
&= -\frac{1}{2} \sum_{j=1}^N \left((\Delta_{\mathbf{r}_j} j(\mathbf{r}_j, \mathbf{r}_p)) + 2 \nabla_{\mathbf{r}_j} j(\mathbf{r}_j, \mathbf{r}_p) \cdot \nabla_{\mathbf{r}_j} \right) \phi(\{\mathbf{r}_i\}, \mathbf{r}_p),
\end{aligned} \tag{19}$$

or that we can write symbolically

$$[\hat{T}_e, J(\{\mathbf{r}_i\}, \mathbf{r}_p)] = -\frac{1}{2} \sum_{j=1}^N \left((\Delta_{\mathbf{r}_j} j(\mathbf{r}_j, \mathbf{r}_p)) + 2 \nabla_{\mathbf{r}_j} j(\mathbf{r}_j, \mathbf{r}_p) \cdot \nabla_{\mathbf{r}_j} \right). \tag{20}$$

Similarly, if one considers the commutator involvin \hat{T}_p one obtains

$$[\hat{T}_p, J(\{\mathbf{r}_i\}, \mathbf{r}_p)] = -\frac{1}{2} \left((\Delta_{\mathbf{r}_p} j(\mathbf{r}_i, \mathbf{r}_p)) + 2 \left[\sum_{i=1}^N \nabla_{\mathbf{r}_p} j(\mathbf{r}_i, \mathbf{r}_p) \right] \cdot \nabla_{\mathbf{r}_p} \right). \tag{21}$$

In these commutators, there is a purely scalar part which is either $\Delta_{\mathbf{r}_j} j(\mathbf{r}_j, \mathbf{r}_p)$ or $\Delta_{\mathbf{r}_p} j(\mathbf{r}_i, \mathbf{r}_p)$ which necessary commutes with the scalar function $J(\{\mathbf{r}_i\}, \mathbf{r}_p)$. Therefore, to compute the second-order commutator, that we divide into two parts

involving the first commutator of \hat{T}_e and that of \hat{T}_p ,

$$\begin{aligned}
\frac{1}{2} \left[[H_{ep}, J(\{\mathbf{r}_i\}, \mathbf{r}_p)], J(\{\mathbf{r}_i\}, \mathbf{r}_p) \right] &= \frac{1}{2} \left[\underbrace{[\hat{T}_e, J(\{\mathbf{r}_i\}, \mathbf{r}_p)], J(\{\mathbf{r}_i\}, \mathbf{r}_p)}_{=C_x+C_y+C_z} \right. \\
&\quad \left. + \frac{1}{2} \left[\underbrace{[\hat{T}_p, J(\{\mathbf{r}_i\}, \mathbf{r}_p)], J(\{\mathbf{r}_i\}, \mathbf{r}_p)}_{=D_x+D_y+D_z} \right] \right]
\end{aligned} \tag{22}$$

only the terms in either $\nabla_{\mathbf{r}_j}$ and $\mathbf{r} \cdot \nabla_{\mathbf{r}_p}$ are not going to commute with $J(\{\mathbf{r}_i\}, \mathbf{r}_p)$. Let us begin C_x

$$\begin{aligned}
C_x &= -\frac{1}{2} \left[\sum_{j=1}^N \frac{\partial}{\partial x_j} j(\mathbf{r}_j, \mathbf{r}_p) \frac{\partial}{\partial x_j}, \sum_{i=1}^N j(\mathbf{r}_i, \mathbf{r}_p) \right] \phi(\{\mathbf{r}_i\}, \mathbf{r}_p) \\
&= -\frac{1}{2} \sum_{j=1}^N \left[\frac{\partial}{\partial x_j} j(\mathbf{r}_j, \mathbf{r}_p) \right] \left(\sum_{i=1}^N \left[\frac{\partial}{\partial x_j} j(\mathbf{r}_i, \mathbf{r}_p) \right] \phi(\{\mathbf{r}_i\}, \mathbf{r}_p) \right) \\
&\quad - \frac{1}{2} \sum_{j=1}^N \left[\frac{\partial}{\partial x_j} j(\mathbf{r}_j, \mathbf{r}_p) \right] \left(\sum_{i=1}^N j(\mathbf{r}_i, \mathbf{r}_p) \right) \frac{\partial}{\partial x_j} \phi(\{\mathbf{r}_i\}, \mathbf{r}_p) \\
&\quad + \frac{1}{2} \sum_{j=1}^N \left[\frac{\partial}{\partial x_j} j(\mathbf{r}_j, \mathbf{r}_p) \right] \left(\sum_{i=1}^N j(\mathbf{r}_i, \mathbf{r}_p) \right) \frac{\partial}{\partial x_j} \phi(\{\mathbf{r}_i\}, \mathbf{r}_p) \\
&= -\frac{1}{2} \sum_{j=1}^N \left[\frac{\partial}{\partial x_j} j(\mathbf{r}_j, \mathbf{r}_p) \right]^2 \phi(\{\mathbf{r}_i\}, \mathbf{r}_p).
\end{aligned} \tag{23}$$

Therefore we can conclude that

$$\frac{1}{2} \left[[\hat{T}_e, J(\{\mathbf{r}_i\}, \mathbf{r}_p)], J(\{\mathbf{r}_i\}, \mathbf{r}_p) \right] = -\sum_{j=1}^N \frac{1}{2} [\nabla_{\mathbf{r}_j} j(\mathbf{r}_j, \mathbf{r}_p)]^2. \tag{24}$$

We then compute the term D

$$\begin{aligned}
D &= -\frac{1}{2} \left[\left[\sum_{i=1}^N \nabla_{\mathbf{r}_p} j(\mathbf{r}_i, \mathbf{r}_p) \right] \cdot \nabla_{\mathbf{r}_p}, \sum_{j=1}^N j(\mathbf{r}_j, \mathbf{r}_p) \right] \phi(\{\mathbf{r}_i\}, \mathbf{r}_p) \\
&= -\frac{1}{2} \sum_{i=1}^N [\nabla_{\mathbf{r}_p} j(\mathbf{r}_i, \mathbf{r}_p)]^2 - \frac{1}{2} \sum_{i=1}^N \sum_{j \neq i} \nabla_{\mathbf{r}_p} j(\mathbf{r}_i, \mathbf{r}_p) \cdot \nabla_{\mathbf{r}_p} j(\mathbf{r}_j, \mathbf{r}_p).
\end{aligned} \tag{25}$$

We can therefore write the total second-order commutator as

$$\begin{aligned}
\frac{1}{2} \left[[\hat{T}, J(\{\mathbf{r}_i\}, \mathbf{r}_p)], J(\{\mathbf{r}_i\}, \mathbf{r}_p) \right] &= \\
&= -\frac{1}{2} \sum_{i=1}^N \left[[\nabla_{\mathbf{r}_p} j(\mathbf{r}_i, \mathbf{r}_p)]^2 + [\nabla_{\mathbf{r}_i} j(\mathbf{r}_i, \mathbf{r}_p)]^2 \right] \\
&\quad - \frac{1}{2} \sum_{i=1}^N \sum_{j \neq i} [\nabla_{\mathbf{r}_p} j(\mathbf{r}_i, \mathbf{r}_p)] \cdot [\nabla_{\mathbf{r}_p} j(\mathbf{r}_j, \mathbf{r}_p)].
\end{aligned} \tag{26}$$

We can therefore write the total transcorrelated Hamiltonian as

$$\begin{aligned}
\tilde{H}_{ep} &= H - \frac{1}{2} \sum_{i=1}^N (\Delta_{\mathbf{r}_i} j(\mathbf{r}_i, \mathbf{r}_p) + \Delta_{\mathbf{r}_p} j(\mathbf{r}_i, \mathbf{r}_p)) \\
&\quad - \sum_{i=1}^N (\nabla_{\mathbf{r}_i} j(\mathbf{r}_i, \mathbf{r}_p) \cdot \nabla_{\mathbf{r}_i} + \nabla_{\mathbf{r}_p} j(\mathbf{r}_i, \mathbf{r}_p) \cdot \nabla_{\mathbf{r}_p}) \\
&\quad - \frac{1}{2} \sum_{i=1}^N [\nabla_{\mathbf{r}_i} j(\mathbf{r}_i, \mathbf{r}_p)]^2 + [\nabla_{\mathbf{r}_p} j(\mathbf{r}_i, \mathbf{r}_p)]^2 \\
&\quad - \frac{1}{2} \sum_{i=1}^N \sum_{j \neq i} [\nabla_{\mathbf{r}_p} j(\mathbf{r}_i, \mathbf{r}_p)] \cdot [\nabla_{\mathbf{r}_p} j(\mathbf{r}_j, \mathbf{r}_p)].
\end{aligned} \tag{27}$$