On the Restricted and Combined Use of Rüdenberg's Approximations in Crystal Orbital Theories of Hartree-Fock Type

Wolfhard Koch[a,b], Bastian Frey[b], Juan Francisco Sánchez Ruiz[a], and Thomas Scior[c]

[a] Universidad Nacional Autónoma de México, Facultad de Estudios Superiores Zaragoza, Av. Guelatao No. 66, Col. Ejército de Oriente, Del. Iztapalapa, 09230 México, D. F., Mexico.
[b] Institut für Physikalische und Theoretische Chemie der Universität Tübingen, Auf der Morgenstelle 8, D-72076 Tübingen, Germany.
[c] Benemérita Universidad Autónoma de Puebla, Facultad de Ciencias Químicas y de Farmacia, 14 Sur con Av. San Claudio, Col. San Miguel, 72570 Puebla, Pue., Mexico.

Reprint requests to Dr. W. K.; Fax: +52 55 5773 6310; E-mail: koch@puma2.zaragoza.unam.mx.

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Dedicated to Prof. Dr. F. F. Seelig on the occasion of his 70th birthday.

The analysis based on Rüdenberg's well-known letter of 1951, which has been outlined for molecules in a preceding contribution, has now been transfered to translational periodic systems in one, two, or three dimensions. Entitled "On the Three- and Four-Center Integrals in Molecular Quantum Mechanics", this letter explicitly presents two approximations only for four-center repulsion integrals. When applied to some types of three-center repulsion integrals, however, these two recipes still imply considerable oversimplifications. Using both one-electron and two-electron routes of Rüdenberg's expansion, such shortcomings can be avoided strictly. Starting from a simple "Unrestricted and Combined" (U&C) approximation scheme introduced elsewhere, an improved "Restricted and Combined" (R&C) approximation picture for Fock-matrix elements now will be outlined, which does not tolerate any unnecessary oversimplifications. Although the simplicity of the U&C scheme is lost in this case, R&C-approximated Fock-matrix elements still can be constructed from one- and two-center integrals alone in an effective way. Moreover, due to their dependence on a single geometric parameter, all types of two-center integrals can be calculated in advance for about one hundred fixed interatomic distances at the desired level of sophistication, and stored once and for all. A cubic spline algorithm may be taken to interpolate the actual integral value from each precomputed list.

Key words: Unrestricted (and Restricted) Hartree-Fock Crystal Orbitals; Integral Approximations According to Rüdenberg; Extended Hückel Theory (EHT).

1. Introduction

As in the preceding contribution [1], the main topic is Rüdenberg's letter of 1951 [2] with its two truncated expansions (symbolized by I and II) of diatomic orbital products. In particular, we consider their application to the crystal-orbital extension of Roothaan's closed-shell "Restricted Hartree-Fock" theory (RHF) [3] in its generalized "Unrestricted Hartree-Fock" form (UHF) of Pople and Nesbet [4].

In analogy to an equivalent molecular orbital study [5], we have already interpreted the consequences of an "Unrestricted and Combined" use (U&C) of Rüdenberg's approximations in crystal-orbital UHF theory [6], apart from any numerical application. Within this context, the term "Unrestricted" indicated that the considered approximations had been applied irrespectively of the one-, two-, and multi-index or -center quality of the integrals involved. (Note that "Unrestricted" in this sense has nothing to do with the conceptual particularities of the "Unrestricted Hartree-Fock" picture itself).

The analysis of [6] yielded the following results:

- The U&C use of Rüdenberg's Mulliken-type approximations [7] (M.U&C) led to a completed understanding of the Wolfsberg-Helmholz formula [8], which is a constituent part of the semi-empirical "Extended Hückel Theory"(EHT) [9].
- Furthermore, an improved Rüdenberg-type variant of M.U&C called R.U&C had been proposed, which is appropriate for non-empirical computer implementations, since it fulfils the "rotational invariance

requirement” [10] of all *ab-initio* quantum chemical concepts.

• Supposing an orthonormal atomic orbital basis set, our M.U&C and R.U&C frameworks immediately converted into two additional “Zero Integral Overlap” (ZIO) and “Neglect of Diatomic Integral Overlap” (NDIO) pictures, which are partially identical with the widely-used ZDO [11] and NDDO [12] approximation schemes, respectively.

• We concluded in pointing out that all these four approximations, which are commonly based on the U&C use of Rüdenberg’s ideas, imply considerable oversimplifications.

In order to overcome such shortcomings, another set of four “Restricted and Combined” (R&C) concepts had been sketched. Here, the attribute “Restricted” indicated the avoidance of particular oversimplifications which is consistent with the corresponding level of approximation.

In the present article we now intend to work out this R&C route of Rüdenberg-type approximations for crystal-orbital theories of Hartree-Fock type. For this purpose it is convenient, first of all, to write down the crystal-orbital UHF Fock-matrix representation in a partitive form which separates explicitly the different one-, two-, three-, and four-center interactions from one another in an appropriate way. In contrast to the preceding molecular version [1] which also included a Mulliken-type, a ZIO, and a NDIO section, the present crystal orbital paper focusses only on the most general branch according to Rüdenberg. It is subdivided into two parts. While the first part intends to explain, how oversimplications generally arise in certain cases of three-index or three-center repulsion integral approximations, the second subsection specifies the “Restricted and Combined” approximation picture for Fock-matrix elements. The equations of these second subsections are constitutive for any forthcoming numerical R.R&C crystal-orbital investigation.

Finally it should be stressed, that such a R&C route of approximations does not improve the quality of all multi-index or multi-center integrals, in general [13]. Improvements, however, can be expected from the fact that conceptual shortcomings of the standard Rüdenberg-picture are minimized in a way, which is designed to be well-balanced in both attractive and repulsive energy contributions.

2. Basic Equations

This chapter is in parts identical with the equivalent one of the R.U&C paper [6]. Leaving out these identical parts including the Pople-Nesbet equations of crystal orbital theory, we therefore immediately continue with the formulation of a real-space Fock-matrix representation, using notation 2.

2.1. Partitive Real-space Fock-matrix Representations Using Notation 2

With a second formulation of the three definitions in ([6] 2.24) we intend to separate explicitly from one another those terms, which represent four-center and three-center interactions. Furthermore, both groups will be isolated from two- or one-center terms.

Introducing the notation $\mathbf{n}N \neq \mathbf{0}M \equiv (\mathbf{n} \neq \mathbf{0}) \vee (N \neq M)$, we write for the off-blockdiagonal attractive part

$$\underbrace{(\mathbf{F}^A_{\mathbf{0n}})_{(M,\mu)(N,\nu)}}_{\mathbf{n}N\neq\mathbf{0}M} = \underbrace{\underbrace{\sum_{\mathbf{p}=\mathbf{N}^-}^{\mathbf{N}^+}\sum_{P=1}^{N_n}}_{\mathbf{p}P\neq\mathbf{0}M,\mathbf{n}N} \begin{pmatrix} \mathbf{0} & \mathbf{p} & \mathbf{n} \\ M & P & N \\ \mu & & \nu \end{pmatrix}}_{\stackrel{\text{def}}{=}({}^{(1)}\mathbf{A}_{\mathbf{0n}})_{(M,\mu)(N,\nu)}} + \underbrace{\begin{pmatrix} \mathbf{0} & \mathbf{n} & \mathbf{n} \\ M & N & N \\ \mu & & \nu \end{pmatrix} + \begin{pmatrix} \mathbf{0} & \mathbf{0} & \mathbf{n} \\ M & M & N \\ \mu & & \nu \end{pmatrix}}_{\stackrel{\text{def}}{=}({}^{(0)}\mathbf{A}_{\mathbf{0n}})_{(M,\mu)(N,\nu)}} . \tag{2.1}$$

For the blockdiagonal attractive part we write

$$(\mathbf{F}^A_{\mathbf{00}})_{(M,\mu)(M,\nu)} = \underbrace{\underbrace{\sum_{\mathbf{p}=\mathbf{N}^-}^{\mathbf{N}^+}\sum_{P=1}^{N_n}}_{\mathbf{p}P\neq\mathbf{0}M} \begin{pmatrix} \mathbf{0} & \mathbf{p} & \mathbf{0} \\ M & P & M \\ \mu & & \nu \end{pmatrix}}_{\stackrel{\text{def}}{=}({}^{(1)}\mathbf{A}_{\mathbf{00}})_{(M,\mu)(M,\nu)}} + \underbrace{\begin{pmatrix} \mathbf{0} & \mathbf{0} & \mathbf{0} \\ M & M & M \\ \mu & & \nu \end{pmatrix}}_{\stackrel{\text{def}}{=}({}^{(0)}\mathbf{A}_{\mathbf{00}})_{(M,\mu)(M,\nu)}} . \tag{2.2}$$

For the off-blockdiagonal Coulomb part we write

$$
\begin{aligned}
\underbrace{(\mathbf{F}^{C}_{\mathbf{0n}})_{(M,\mu)(N,\nu)}}_{\mathbf{n}N\neq\mathbf{0}M} = {} & \underbrace{\underbrace{\sum_{\mathbf{t}=\mathbf{N}^-}^{\mathbf{N}^+}\sum_{T=1}^{N_n}}_{\mathbf{t}T\neq\mathbf{0}M,\mathbf{n}N}\sum_{\tau=1}^{n_o(T)}\underbrace{\sum_{\mathbf{l}=\mathbf{N}^-}^{\mathbf{N}^+}\sum_{L=1}^{N_n}}_{\mathbf{l}L\neq\mathbf{0}M,\mathbf{n}N,\mathbf{t}T}\sum_{\lambda=1}^{n_o(L)}(\mathbf{P}^{\oplus}_{\mathbf{tl}})_{(T,\tau)(L,\lambda)}\left(\begin{array}{cc|cc}\mathbf{0}&\mathbf{n}&\mathbf{t}&\mathbf{l}\\ M&N&T&L\\ \mu&\nu&\tau&\lambda\end{array}\right)}_{\overset{\text{def}}{=}({}^{(1)}\mathbf{C}_{\mathbf{0n}})_{(M,\mu)(N,\nu)}}\\
& + \underbrace{\underbrace{\sum_{\mathbf{t}=\mathbf{N}^-}^{\mathbf{N}^+}\sum_{T=1}^{N_n}}_{\mathbf{t}T\neq\mathbf{0}M,\mathbf{n}N}\sum_{\tau=1}^{n_o(T)}\sum_{\lambda=1}^{n_o(T)}(\mathbf{P}^{\oplus}_{\mathbf{tt}})_{(T,\tau)(T,\lambda)}\left(\begin{array}{cc|cc}\mathbf{0}&\mathbf{n}&\mathbf{t}&\mathbf{t}\\ M&N&T&T\\ \mu&\nu&\tau&\lambda\end{array}\right)}_{\overset{\text{def}}{=}({}^{(2)}\mathbf{C}_{\mathbf{0n}})_{(M,\mu)(N,\nu)}}\\
& + \underbrace{2\underbrace{\sum_{\mathbf{t}=\mathbf{N}^-}^{\mathbf{N}^+}\sum_{T=1}^{N_n}}_{\mathbf{t}T\neq\mathbf{0}M,\mathbf{n}N}\sum_{\tau=1}^{n_o(T)}\sum_{\lambda=1}^{n_o(M)}(\mathbf{P}^{\oplus}_{\mathbf{t0}})_{(T,\tau)(M,\lambda)}\left(\begin{array}{cc|cc}\mathbf{0}&\mathbf{n}&\mathbf{t}&\mathbf{0}\\ M&N&T&M\\ \mu&\nu&\tau&\lambda\end{array}\right)}_{\overset{\text{def}}{=}({}^{(3)}\mathbf{C}_{\mathbf{0n}})_{(M,\mu)(N,\nu)}}\\
& + \underbrace{2\underbrace{\sum_{\mathbf{t}=\mathbf{N}^-}^{\mathbf{N}^+}\sum_{T=1}^{N_n}}_{\mathbf{t}T\neq\mathbf{0}M,\mathbf{n}N}\sum_{\tau=1}^{n_o(T)}\sum_{\lambda=1}^{n_o(N)}(\mathbf{P}^{\oplus}_{\mathbf{tn}})_{(T,\tau)(N,\lambda)}\left(\begin{array}{cc|cc}\mathbf{0}&\mathbf{n}&\mathbf{t}&\mathbf{n}\\ M&N&T&N\\ \mu&\nu&\tau&\lambda\end{array}\right)}_{\overset{\text{def}}{=}({}^{(4)}\mathbf{C}_{\mathbf{0n}})_{(M,\mu)(N,\nu)}} + ({}^{(0)}\mathbf{C}_{\mathbf{0n}})_{(M,\mu)(N,\nu)}
\end{aligned}
\tag{2.3}
$$

with

$$
\begin{aligned}
\underbrace{({}^{(0)}\mathbf{C}_{\mathbf{0n}})_{(M,\mu)(N,\nu)}}_{\mathbf{n}N\neq\mathbf{0}M} \overset{\text{def}}{=} {} & \sum_{\tau=1}^{n_o(M)}\sum_{\lambda=1}^{n_o(M)}(\mathbf{P}^{\oplus}_{\mathbf{00}})_{(M,\tau)(M,\lambda)}\left(\begin{array}{cc|cc}\mathbf{0}&\mathbf{n}&\mathbf{0}&\mathbf{0}\\ M&N&M&M\\ \mu&\nu&\tau&\lambda\end{array}\right)\\
& + \sum_{\tau=1}^{n_o(N)}\sum_{\lambda=1}^{n_o(N)}(\mathbf{P}^{\oplus}_{\mathbf{00}})_{(N,\tau)(N,\lambda)}\left(\begin{array}{cc|cc}\mathbf{0}&\mathbf{n}&\mathbf{n}&\mathbf{n}\\ M&N&N&N\\ \mu&\nu&\tau&\lambda\end{array}\right)\\
& + 2\sum_{\tau=1}^{n_o(M)}\sum_{\lambda=1}^{n_o(N)}(\mathbf{P}^{\oplus}_{\mathbf{0n}})_{(M,\tau)(N,\lambda)}\left(\begin{array}{cc|cc}\mathbf{0}&\mathbf{n}&\mathbf{0}&\mathbf{n}\\ M&N&M&N\\ \mu&\nu&\tau&\lambda\end{array}\right).
\end{aligned}
\tag{2.4}
$$

For the blockdiagonal Coulomb part we write

$$
(\mathbf{F}^{C}_{\mathbf{00}})_{(M,\mu)(M,\nu)} = \underbrace{\underbrace{\sum_{\mathbf{t}=\mathbf{N}^-}^{\mathbf{N}^+}\sum_{T=1}^{N_n}}_{\mathbf{t}T\neq\mathbf{0}M}\sum_{\tau=1}^{n_o(T)}\underbrace{\sum_{\mathbf{l}=\mathbf{N}^-}^{\mathbf{N}^+}\sum_{L=1}^{N_n}}_{\mathbf{l}L\neq\mathbf{0}M,\mathbf{t}T}\sum_{\lambda=1}^{n_o(L)}(\mathbf{P}^{\oplus}_{\mathbf{tl}})_{(T,\tau)(L,\lambda)}\left(\begin{array}{cc|cc}\mathbf{0}&\mathbf{0}&\mathbf{t}&\mathbf{l}\\ M&M&T&L\\ \mu&\nu&\tau&\lambda\end{array}\right)}_{\overset{\text{def}}{=}({}^{(1)}\mathbf{C}_{\mathbf{00}})_{(M,\mu)(M,\nu)}}
$$

$$
\begin{aligned}
&+ \underbrace{\underbrace{\sum_{\mathbf{t}=\mathbf{N}^-}^{\mathbf{N}^+} \sum_{T=1}^{N_n}}_{\mathbf{t}T\neq\mathbf{0}M} \sum_{\tau=1}^{n_o(T)} \sum_{\lambda=1}^{n_o(T)} (\mathbf{P}^{\oplus}_{\mathbf{00}})_{(T,\tau)(T,\lambda)} \left(\begin{array}{cc|cc} \mathbf{0} & \mathbf{0} & \mathbf{t} & \mathbf{t} \\ M & M & T & T \\ \mu & \nu & \tau & \lambda \end{array}\right)}_{\stackrel{\text{def}}{=}({}^{(2)}\mathbf{C}_{\mathbf{00}})_{(M,\mu)(M,\nu)}} \\
&+ \underbrace{2 \underbrace{\sum_{\mathbf{t}=\mathbf{N}^-}^{\mathbf{N}^+} \sum_{T=1}^{N_n}}_{\mathbf{t}T\neq\mathbf{0}M} \sum_{\tau=1}^{n_o(T)} \sum_{\lambda=1}^{n_o(M)} (\mathbf{P}^{\oplus}_{\mathbf{0t}})_{(M,\lambda)(T,\tau)} \left(\begin{array}{cc|cc} \mathbf{0} & \mathbf{0} & \mathbf{t} & \mathbf{0} \\ M & M & T & M \\ \mu & \nu & \tau & \lambda \end{array}\right)}_{\stackrel{\text{def}}{=}({}^{(3)}\mathbf{C}_{\mathbf{00}})_{(M,\mu)(M,\nu)}} \qquad (2.5) \\
&+ \underbrace{\sum_{\tau=1}^{n_o(M)} \sum_{\lambda=1}^{n_o(M)} (\mathbf{P}^{\oplus}_{\mathbf{00}})_{(M,\tau)(M,\lambda)} \left(\begin{array}{cc|cc} \mathbf{0} & \mathbf{0} & \mathbf{0} & \mathbf{0} \\ M & M & M & M \\ \mu & \nu & \tau & \lambda \end{array}\right)}_{\stackrel{\text{def}}{=}({}^{(0)}\mathbf{C}_{\mathbf{00}})_{(M,\mu)(M,\nu)}} .
\end{aligned}
$$

For the off-blockdiagonal exchange part we write

$$
\begin{aligned}
\underbrace{(\mathbf{F}^{\alpha E}_{\mathbf{0n}})_{(M,\mu)(N,\nu)}}_{\mathbf{n}N\neq\mathbf{0}M} &= \underbrace{\underbrace{\sum_{\mathbf{t}=\mathbf{N}^-}^{\mathbf{N}^+} \sum_{T=1}^{N_n}}_{\mathbf{t}T\neq\mathbf{0}M,\mathbf{n}N} \sum_{\tau=1}^{n_o(T)} \underbrace{\sum_{\mathbf{l}=\mathbf{N}^-}^{\mathbf{N}^+} \sum_{L=1}^{N_n}}_{\mathbf{l}L\neq\mathbf{0}M,\mathbf{n}N,\mathbf{t}T} \sum_{\lambda=1}^{n_o(L)} (\mathbf{P}^{\alpha}_{\mathbf{tl}})_{(T,\tau)(L,\lambda)} \left(\begin{array}{cc|cc} \mathbf{0} & \mathbf{t} & \mathbf{n} & \mathbf{l} \\ M & T & N & L \\ \mu & \tau & \nu & \lambda \end{array}\right)}_{\stackrel{\text{def}}{=}({}^{(1)}\mathbf{E}^{\alpha}_{\mathbf{0n}})_{(M,\mu)(N,\nu)}} \\
&+ \underbrace{\underbrace{\sum_{\mathbf{t}=\mathbf{N}^-}^{\mathbf{N}^+} \sum_{T=1}^{N_n}}_{\mathbf{t}T\neq\mathbf{0}M,\mathbf{n}N} \sum_{\tau=1}^{n_o(T)} \sum_{\lambda=1}^{n_o(T)} (\mathbf{P}^{\alpha}_{\mathbf{tt}})_{(T,\tau)(T,\lambda)} \left(\begin{array}{cc|cc} \mathbf{0} & \mathbf{t} & \mathbf{n} & \mathbf{t} \\ M & T & N & T \\ \mu & \tau & \nu & \lambda \end{array}\right)}_{\stackrel{\text{def}}{=}({}^{(2)}\mathbf{E}^{\alpha}_{\mathbf{0n}})_{(M,\mu)(N,\nu)}} \\
&+ \underbrace{\underbrace{\sum_{\mathbf{t}=\mathbf{N}^-}^{\mathbf{N}^+} \sum_{T=1}^{N_n}}_{\mathbf{t}T\neq\mathbf{0}M,\mathbf{n}N} \sum_{\tau=1}^{n_o(T)} \sum_{\lambda=1}^{n_o(M)} (\mathbf{P}^{\alpha}_{\mathbf{t0}})_{(T,\tau)(M,\lambda)} \left(\begin{array}{cc|cc} \mathbf{0} & \mathbf{t} & \mathbf{n} & \mathbf{0} \\ M & T & N & M \\ \mu & \tau & \nu & \lambda \end{array}\right)}_{\stackrel{\text{def}}{=}({}^{(3)}\mathbf{E}^{\alpha}_{\mathbf{0n}})_{(M,\mu)(N,\nu)}} \\
&+ \underbrace{\underbrace{\sum_{\mathbf{t}=\mathbf{N}^-}^{\mathbf{N}^+} \sum_{T=1}^{N_n}}_{\mathbf{t}T\neq\mathbf{0}M,\mathbf{n}N} \sum_{\tau=1}^{n_o(T)} \sum_{\lambda=1}^{n_o(N)} (\mathbf{P}^{\alpha}_{\mathbf{tn}})_{(T,\tau)(N,\lambda)} \left(\begin{array}{cc|cc} \mathbf{0} & \mathbf{t} & \mathbf{n} & \mathbf{n} \\ M & T & N & N \\ \mu & \tau & \nu & \lambda \end{array}\right)}_{\stackrel{\text{def}}{=}({}^{(4)}\mathbf{E}^{\alpha}_{\mathbf{0n}})_{(M,\mu)(N,\nu)}}
\end{aligned}
$$

$$
\begin{aligned}
&+\underbrace{\underbrace{\sum_{\mathbf{l}=\mathbf{N}^-}^{\mathbf{N}^+}\sum_{L=1}^{N_n}}_{\mathbf{l}L\neq\mathbf{0}M,\mathbf{n}N}\sum_{\tau=1}^{n_o(M)}\sum_{\lambda=1}^{n_o(L)}(\mathbf{P}^{\alpha}_{\mathbf{0l}})_{(M,\tau)(L,\lambda)}\left(\begin{array}{cc|cc}\mathbf{0}&\mathbf{0}&\mathbf{n}&\mathbf{l}\\ M&M&N&L\\ \mu&\tau&\nu&\lambda\end{array}\right)}_{\stackrel{\text{def}}{=}({}^{(5)}\mathbf{E}^{\alpha}_{\mathbf{0n}})_{(M,\mu)(N,\nu)}}\\
&+\underbrace{\underbrace{\sum_{\mathbf{l}=\mathbf{N}^-}^{\mathbf{N}^+}\sum_{L=1}^{N_n}}_{\mathbf{l}L\neq\mathbf{0}M,\mathbf{n}N}\sum_{\tau=1}^{n_o(N)}\sum_{\lambda=1}^{n_o(L)}(\mathbf{P}^{\alpha}_{\mathbf{nl}})_{(N,\tau)(L,\lambda)}\left(\begin{array}{cc|cc}\mathbf{0}&\mathbf{n}&\mathbf{n}&\mathbf{l}\\ M&N&N&L\\ \mu&\tau&\nu&\lambda\end{array}\right)}_{\stackrel{\text{def}}{=}({}^{(6)}\mathbf{E}^{\alpha}_{\mathbf{0n}})_{(M,\mu)(N,\nu)}}+({}^{(0)}\mathbf{E}^{\alpha}_{\mathbf{0n}})_{(M,\mu)(N,\nu)}
\end{aligned}
\tag{2.6}
$$

with

$$
\begin{aligned}
\underbrace{({}^{(0)}\mathbf{E}^{\alpha}_{\mathbf{0n}})_{(M,\mu)(N,\nu)}}_{\mathbf{n}N\neq\mathbf{0}M}\stackrel{\text{def}}{=}&\sum_{\tau=1}^{n_o(M)}\sum_{\lambda=1}^{n_o(M)}(\mathbf{P}^{\alpha}_{\mathbf{00}})_{(M,\tau)(M,\lambda)}\left(\begin{array}{cc|cc}\mathbf{0}&\mathbf{0}&\mathbf{n}&\mathbf{0}\\ M&M&N&M\\ \mu&\tau&\nu&\lambda\end{array}\right)+\sum_{\tau=1}^{n_o(N)}\sum_{\lambda=1}^{n_o(N)}(\mathbf{P}^{\alpha}_{\mathbf{00}})_{(N,\tau)(N,\lambda)}\left(\begin{array}{cc|cc}\mathbf{0}&\mathbf{n}&\mathbf{n}&\mathbf{n}\\ M&N&N&N\\ \mu&\tau&\nu&\lambda\end{array}\right)\\
&+\sum_{\tau=1}^{n_o(M)}\sum_{\lambda=1}^{n_o(N)}(\mathbf{P}^{\alpha}_{\mathbf{0n}})_{(M,\tau)(N,\lambda)}\left(\begin{array}{cc|cc}\mathbf{0}&\mathbf{0}&\mathbf{n}&\mathbf{n}\\ M&M&N&N\\ \mu&\tau&\nu&\lambda\end{array}\right)+\sum_{\tau=1}^{n_o(N)}\sum_{\lambda=1}^{n_o(M)}(\mathbf{P}^{\alpha}_{\mathbf{0n}})_{(M,\lambda)(N,\tau)}\left(\begin{array}{cc|cc}\mathbf{0}&\mathbf{n}&\mathbf{n}&\mathbf{0}\\ M&N&N&M\\ \mu&\tau&\nu&\lambda\end{array}\right).
\end{aligned}
\tag{2.7}
$$

For the blockdiagonal exchange part we write

$$
\begin{aligned}
(\mathbf{F}^{\alpha E}_{\mathbf{00}})_{(M,\mu)(M,\nu)}=&\underbrace{\underbrace{\sum_{\mathbf{t}=\mathbf{N}^-}^{\mathbf{N}^+}\sum_{T=1}^{N_n}}_{\mathbf{t}T\neq\mathbf{0}M}\sum_{\tau=1}^{n_o(T)}\underbrace{\sum_{\mathbf{l}=\mathbf{N}^-}^{\mathbf{N}^+}\sum_{L=1}^{N_n}}_{\mathbf{l}L\neq\mathbf{0}M,\mathbf{t}T}\sum_{\lambda=1}^{n_o(L)}(\mathbf{P}^{\alpha}_{\mathbf{tl}})_{(T,\tau)(L,\lambda)}\left(\begin{array}{cc|cc}\mathbf{0}&\mathbf{t}&\mathbf{0}&\mathbf{l}\\ M&T&M&L\\ \mu&\tau&\nu&\lambda\end{array}\right)}_{\stackrel{\text{def}}{=}({}^{(1)}\mathbf{E}^{\alpha}_{\mathbf{00}})_{(M,\mu)(M,\nu)}}\\
&+\underbrace{\underbrace{\sum_{\mathbf{t}=\mathbf{N}^-}^{\mathbf{N}^+}\sum_{T=1}^{N_n}}_{\mathbf{t}T\neq\mathbf{0}M}\sum_{\tau=1}^{n_o(T)}\sum_{\lambda=1}^{n_o(T)}(\mathbf{P}^{\alpha}_{\mathbf{00}})_{(T,\tau)(T,\lambda)}\left(\begin{array}{cc|cc}\mathbf{0}&\mathbf{t}&\mathbf{0}&\mathbf{t}\\ M&T&M&T\\ \mu&\tau&\nu&\lambda\end{array}\right)}_{\stackrel{\text{def}}{=}({}^{(2)}\mathbf{E}^{\alpha}_{\mathbf{00}})_{(M,\mu)(M,\nu)}}+\underbrace{\underbrace{\sum_{\mathbf{t}=\mathbf{N}^-}^{\mathbf{N}^+}\sum_{T=1}^{N_n}}_{\mathbf{t}T\neq\mathbf{0}M}\sum_{\tau=1}^{n_o(T)}\sum_{\lambda=1}^{n_o(M)}(\mathbf{P}^{\alpha}_{\mathbf{0t}})_{(M,\lambda)(T,\tau)}\left(\begin{array}{cc|cc}\mathbf{0}&\mathbf{t}&\mathbf{0}&\mathbf{0}\\ M&T&M&M\\ \mu&\tau&\nu&\lambda\end{array}\right)}_{\stackrel{\text{def}}{=}({}^{(3)}\mathbf{E}^{\alpha}_{\mathbf{00}})_{(M,\mu)(M,\nu)}}\\
&+\underbrace{\underbrace{\sum_{\mathbf{l}=\mathbf{N}^-}^{\mathbf{N}^+}\sum_{L=1}^{N_n}}_{\mathbf{l}L\neq\mathbf{0}M}\sum_{\lambda=1}^{n_o(L)}\sum_{\tau=1}^{n_o(M)}(\mathbf{P}^{\alpha}_{\mathbf{0l}})_{(M,\tau)(L,\lambda)}\left(\begin{array}{cc|cc}\mathbf{0}&\mathbf{0}&\mathbf{0}&\mathbf{l}\\ M&M&M&L\\ \mu&\tau&\nu&\lambda\end{array}\right)}_{\stackrel{\text{def}}{=}({}^{(5)}\mathbf{E}^{\alpha}_{\mathbf{00}})_{(M,\mu)(M,\nu)}}+\underbrace{\sum_{\tau=1}^{n_o(M)}\sum_{\lambda=1}^{n_o(M)}(\mathbf{P}^{\alpha}_{\mathbf{00}})_{(M,\tau)(M,\lambda)}\left(\begin{array}{cc|cc}\mathbf{0}&\mathbf{0}&\mathbf{0}&\mathbf{0}\\ M&M&M&M\\ \mu&\tau&\nu&\lambda\end{array}\right)}_{\stackrel{\text{def}}{=}({}^{(0)}\mathbf{E}^{\alpha}_{\mathbf{00}})_{(M,\mu)(M,\nu)}}.
\end{aligned}
\tag{2.8}
$$

It should be stressed once again that the ensemble of partitive formulations of (2.1) … (2.8) are completely equivalent to the compact formulations of ([6] 2.24).

2.2. Approximations for Integrals and Fock-matrix Elements

Since reciprocal-space integrations according to Ramírez and Böhm [14] have already been described in the preceding crystal orbital paper, we now turn to the discussion of an approximation method based on Rüdenberg's ideas contained in his famous short paper of 1951 [2] entitled "*On the Three- and Four-Center Integrals in Molecular Quantum Mechanics*". Obviously, this title suggests that Rüdenberg does not recommend his approximation for all types of two-center integrals. When applied on certain three-center repulsion integrals, however, his recipe still implies considerable oversimplifications, as we shall see, which have not been discussed explicitly in his contribution. Using the one- and two-electron routes of Rüdenberg's expansion, on the other hand, these shortcomings can be strictly avoided.

Besides the integral approximations themselves, our interest is focussed on their effect for the evaluation of matrix elements occuring in the UHF representation.

3. Approximations of Rüdenberg Type

3.1. "Unrestricted" and "Restricted" Integral Approximations

In particular, Rüdenberg's approximation intends to reduce the four-center repulsion integrals to those of two-center type. According to his letter of 1951, this aim can be reached in two ways. The first (standard) approach consists in expanding a differential two-center one-electron density as follows:

$$\text{(I)}\quad \left\{\boldsymbol{\Phi}_\mu(\mathbf{r}_i-\mathbf{R}_M-\mathbf{R}_\mathbf{m})\boldsymbol{\Phi}_\nu(\mathbf{r}_i-\mathbf{R}_N-\mathbf{R}_\mathbf{n})\right\}^{[\mathrm{R}^{\mathrm{I}}_{MN}]} := \tag{3.1}$$

$$\frac{1}{2}\left\{\sum_{\mu'=1}^{n_o(M)}\begin{pmatrix}\mathbf{m} & \mathbf{n}\\ M & N\\ \mu' & \nu\end{pmatrix}\boldsymbol{\Phi}_\mu(\mathbf{r}_i-\mathbf{R}_M-\mathbf{R}_\mathbf{m})\boldsymbol{\Phi}_{\mu'}(\mathbf{r}_i-\mathbf{R}_M-\mathbf{R}_\mathbf{m})+\sum_{\nu'=1}^{n_o(N)}\begin{pmatrix}\mathbf{m} & \mathbf{n}\\ M & N\\ \mu & \nu'\end{pmatrix}\boldsymbol{\Phi}_\nu(\mathbf{r}_i-\mathbf{R}_N-\mathbf{R}_\mathbf{n})\boldsymbol{\Phi}_{\nu'}(\mathbf{r}_i-\mathbf{R}_N-\mathbf{R}_\mathbf{n})\right\}.$$

Alternatively, one can also impose such an expansion on six-dimensional two-center two-electron orbital products:

$$\text{(II)}\quad \left\{\boldsymbol{\Phi}_\mu(\mathbf{r}_i-\mathbf{R}_M-\mathbf{R}_\mathbf{m})\boldsymbol{\Phi}_\nu(\mathbf{r}_j-\mathbf{R}_N-\mathbf{R}_\mathbf{n})\right\}^{[\mathrm{R}^{\mathrm{II}}_{MN}]} \tag{3.2}$$

$$\frac{1}{2}\left\{\sum_{\mu'=1}^{n_o(M)}\begin{pmatrix}\mathbf{m} & \mathbf{n}\\ M & N\\ \mu' & \nu\end{pmatrix}\boldsymbol{\Phi}_\mu(\mathbf{r}_i-\mathbf{R}_M-\mathbf{R}_\mathbf{m})\boldsymbol{\Phi}_{\mu'}(\mathbf{r}_j-\mathbf{R}_M-\mathbf{R}_\mathbf{m})+\sum_{\nu'=1}^{n_o(N)}\begin{pmatrix}\mathbf{m} & \mathbf{n}\\ M & N\\ \mu & \nu'\end{pmatrix}\boldsymbol{\Phi}_\nu(\mathbf{r}_i-\mathbf{R}_N-\mathbf{R}_\mathbf{n})\boldsymbol{\Phi}_{\nu'}(\mathbf{r}_j-\mathbf{R}_N-\mathbf{R}_\mathbf{n})\right\}.$$

In a Rüdenberg-type treatment of four-center repulsion integrals each of both routes has to be passed twice:

$$\begin{aligned}
\text{(I)}\quad &\left(\begin{array}{cc|cc}\mathbf{m} & \mathbf{n} & \mathbf{t} & \mathbf{l}\\ M & N & T & L\\ \mu & \nu & \tau & \lambda\end{array}\right)^{[\mathrm{R}^{\mathrm{I}}_{MN}\mathrm{R}^{\mathrm{I}}_{TL}]} := \frac{1}{2}\left\{\sum_{\mu'=1}^{n_o(M)}\begin{pmatrix}\mathbf{m} & \mathbf{n}\\ M & N\\ \mu' & \nu\end{pmatrix}\left(\begin{array}{cc|cc}\mathbf{m} & \mathbf{m} & \mathbf{t} & \mathbf{l}\\ M & M & T & L\\ \mu & \mu' & \tau & \lambda\end{array}\right)^{[\mathrm{R}^{\mathrm{I}}_{TL}]}+\sum_{\nu'=1}^{n_o(N)}\begin{pmatrix}\mathbf{m} & \mathbf{n}\\ M & N\\ \mu & \nu'\end{pmatrix}\left(\begin{array}{cc|cc}\mathbf{n} & \mathbf{n} & \mathbf{t} & \mathbf{l}\\ N & N & T & L\\ \nu' & \nu & \tau & \lambda\end{array}\right)^{[\mathrm{R}^{\mathrm{I}}_{TL}]}\right\}\\
&:= \frac{1}{4}\left\{\sum_{\mu'=1}^{n_o(M)}\begin{pmatrix}\mathbf{m} & \mathbf{n}\\ M & N\\ \mu' & \nu\end{pmatrix}\left[\sum_{\tau'=1}^{n_o(T)}\begin{pmatrix}\mathbf{t} & \mathbf{l}\\ T & L\\ \tau' & \lambda\end{pmatrix}\left(\begin{array}{cc|cc}\mathbf{m} & \mathbf{m} & \mathbf{t} & \mathbf{t}\\ M & M & T & T\\ \mu & \mu' & \tau & \tau'\end{array}\right)+\sum_{\lambda'=1}^{n_o(L)}\begin{pmatrix}\mathbf{t} & \mathbf{l}\\ T & L\\ \tau & \lambda'\end{pmatrix}\left(\begin{array}{cc|cc}\mathbf{m} & \mathbf{m} & \mathbf{l} & \mathbf{l}\\ M & M & L & L\\ \mu & \mu' & \lambda' & \lambda\end{array}\right)\right]\right.\\
&\qquad\left.+\sum_{\nu'=1}^{n_o(N)}\begin{pmatrix}\mathbf{m} & \mathbf{n}\\ M & N\\ \mu & \nu'\end{pmatrix}\left[\sum_{\tau'=1}^{n_o(T)}\begin{pmatrix}\mathbf{t} & \mathbf{l}\\ T & L\\ \tau' & \lambda\end{pmatrix}\left(\begin{array}{cc|cc}\mathbf{n} & \mathbf{n} & \mathbf{t} & \mathbf{t}\\ N & N & T & T\\ \nu' & \nu & \tau & \tau'\end{array}\right)+\sum_{\lambda'=1}^{n_o(L)}\begin{pmatrix}\mathbf{t} & \mathbf{l}\\ T & L\\ \tau & \lambda'\end{pmatrix}\left(\begin{array}{cc|cc}\mathbf{n} & \mathbf{n} & \mathbf{l} & \mathbf{l}\\ N & N & L & L\\ \nu' & \nu & \lambda' & \lambda\end{array}\right)\right]\right\},
\end{aligned}\tag{3.3}$$

$$\begin{aligned}
\text{(II)}\quad &\left(\begin{array}{cc|cc}\mathbf{m} & \mathbf{n} & \mathbf{t} & \mathbf{l}\\ M & N & T & L\\ \mu & \nu & \tau & \lambda\end{array}\right)^{[\mathrm{R}^{\mathrm{II}}_{MT}\mathrm{R}^{\mathrm{II}}_{NL}]} := \frac{1}{2}\left\{\sum_{\mu'=1}^{n_o(M)}\begin{pmatrix}\mathbf{m} & \mathbf{t}\\ M & T\\ \mu' & \tau\end{pmatrix}\left(\begin{array}{cc|cc}\mathbf{m} & \mathbf{n} & \mathbf{m} & \mathbf{l}\\ M & N & M & L\\ \mu & \nu & \mu' & \lambda\end{array}\right)^{[\mathrm{R}^{\mathrm{II}}_{NL}]}+\sum_{\tau'=1}^{n_o(T)}\begin{pmatrix}\mathbf{m} & \mathbf{t}\\ M & T\\ \mu & \tau'\end{pmatrix}\left(\begin{array}{cc|cc}\mathbf{t} & \mathbf{n} & \mathbf{t} & \mathbf{l}\\ T & N & T & L\\ \tau' & \nu & \tau & \lambda\end{array}\right)^{[\mathrm{R}^{\mathrm{II}}_{NL}]}\right\}\\
&:= \frac{1}{4}\left\{\sum_{\mu'=1}^{n_o(M)}\begin{pmatrix}\mathbf{m} & \mathbf{t}\\ M & T\\ \mu' & \tau\end{pmatrix}\left[\sum_{\nu'=1}^{n_o(N)}\begin{pmatrix}\mathbf{n} & \mathbf{l}\\ N & L\\ \nu' & \lambda\end{pmatrix}\left(\begin{array}{cc|cc}\mathbf{m} & \mathbf{n} & \mathbf{m} & \mathbf{n}\\ M & N & M & N\\ \mu & \nu & \mu' & \nu'\end{array}\right)+\sum_{\lambda'=1}^{n_o(L)}\begin{pmatrix}\mathbf{n} & \mathbf{l}\\ N & L\\ \nu & \lambda'\end{pmatrix}\left(\begin{array}{cc|cc}\mathbf{m} & \mathbf{l} & \mathbf{m} & \mathbf{l}\\ M & L & M & L\\ \mu & \lambda' & \mu' & \lambda\end{array}\right)\right]\right.\\
&\qquad\left.+\sum_{\tau'=1}^{n_o(T)}\begin{pmatrix}\mathbf{m} & \mathbf{t}\\ M & T\\ \mu & \tau'\end{pmatrix}\left[\sum_{\nu'=1}^{n_o(N)}\begin{pmatrix}\mathbf{n} & \mathbf{l}\\ N & L\\ \nu' & \lambda\end{pmatrix}\left(\begin{array}{cc|cc}\mathbf{t} & \mathbf{n} & \mathbf{t} & \mathbf{n}\\ T & N & T & N\\ \tau' & \nu & \tau & \nu'\end{array}\right)+\sum_{\lambda'=1}^{n_o(L)}\begin{pmatrix}\mathbf{n} & \mathbf{l}\\ N & L\\ \nu & \lambda'\end{pmatrix}\left(\begin{array}{cc|cc}\mathbf{t} & \mathbf{l} & \mathbf{t} & \mathbf{l}\\ T & L & T & L\\ \tau' & \lambda' & \tau & \lambda\end{array}\right)\right]\right\}.
\end{aligned}\tag{3.4}$$

Having interchanged $\mathbf{n}$, N and ν with $\mathbf{t}$, T and τ, respectively, (3.4) equivalently reads

$$
\begin{aligned}
\text{(II)}\quad \left(\begin{array}{cc|cc}\mathbf{m}&\mathbf{t}&\mathbf{n}&\mathbf{l}\\ M&T&N&L\\ \mu&\tau&\nu&\lambda\end{array}\right)^{[\mathrm{R}^{\mathrm{II}}_{MN}\mathrm{R}^{\mathrm{II}}_{TL}]} &:= \frac{1}{2}\Bigg\{\sum_{\mu'=1}^{n_o(M)}\begin{pmatrix}\mathbf{m}&\mathbf{n}\\ M&N\\ \mu'&\nu\end{pmatrix}\left(\begin{array}{cc|cc}\mathbf{m}&\mathbf{t}&\mathbf{m}&\mathbf{l}\\ M&T&M&L\\ \mu&\tau&\mu'&\lambda\end{array}\right)^{[\mathrm{R}^{\mathrm{II}}_{TL}]} + \sum_{\nu'=1}^{n_o(N)}\begin{pmatrix}\mathbf{m}&\mathbf{n}\\ M&N\\ \mu&\nu'\end{pmatrix}\left(\begin{array}{cc|cc}\mathbf{n}&\mathbf{t}&\mathbf{n}&\mathbf{l}\\ N&T&N&L\\ \nu'&\tau&\nu&\lambda\end{array}\right)^{[\mathrm{R}^{\mathrm{II}}_{TL}]}\Bigg\}\\
&:= \frac{1}{4}\Bigg\{\sum_{\mu'=1}^{n_o(M)}\begin{pmatrix}\mathbf{m}&\mathbf{n}\\ M&N\\ \mu'&\nu\end{pmatrix}\Bigg[\sum_{\tau'=1}^{n_o(T)}\begin{pmatrix}\mathbf{t}&\mathbf{l}\\ T&L\\ \tau'&\lambda\end{pmatrix}\left(\begin{array}{cc|cc}\mathbf{m}&\mathbf{t}&\mathbf{m}&\mathbf{t}\\ M&T&M&T\\ \mu&\tau&\mu'&\tau'\end{array}\right) + \sum_{\lambda'=1}^{n_o(L)}\begin{pmatrix}\mathbf{t}&\mathbf{l}\\ T&L\\ \tau&\lambda'\end{pmatrix}\left(\begin{array}{cc|cc}\mathbf{m}&\mathbf{l}&\mathbf{m}&\mathbf{l}\\ M&L&M&L\\ \mu&\lambda'&\mu'&\lambda\end{array}\right)\Bigg]\\
&\quad + \sum_{\nu'=1}^{n_o(N)}\begin{pmatrix}\mathbf{m}&\mathbf{n}\\ M&N\\ \mu&\nu'\end{pmatrix}\Bigg[\sum_{\tau'=1}^{n_o(T)}\begin{pmatrix}\mathbf{t}&\mathbf{l}\\ T&L\\ \tau'&\lambda\end{pmatrix}\left(\begin{array}{cc|cc}\mathbf{n}&\mathbf{t}&\mathbf{n}&\mathbf{t}\\ N&T&N&T\\ \nu'&\tau&\nu&\tau'\end{array}\right) + \sum_{\lambda'=1}^{n_o(L)}\begin{pmatrix}\mathbf{t}&\mathbf{l}\\ T&L\\ \tau&\lambda'\end{pmatrix}\left(\begin{array}{cc|cc}\mathbf{n}&\mathbf{l}&\mathbf{n}&\mathbf{l}\\ N&L&N&L\\ \nu'&\lambda'&\nu&\lambda\end{array}\right)\Bigg]\Bigg\}.
\end{aligned} \tag{3.5}
$$

In addition to these formulas, which are already contained in Rüdenberg's letter, let us consider the three-center repulsion integrals $\left(\begin{array}{cc|cc}\mathbf{m}&\mathbf{m}&\mathbf{t}&\mathbf{l}\\ M&M&T&L\\ \mu&\nu&\tau&\lambda\end{array}\right)$ and $\left(\begin{array}{cc|cc}\mathbf{m}&\mathbf{t}&\mathbf{m}&\mathbf{l}\\ M&T&M&L\\ \mu&\tau&\nu&\lambda\end{array}\right)$.

(I) Using two one-electron approximations of Rüdenberg type, we get

$$
\begin{aligned}
\left(\begin{array}{cc|cc}\mathbf{m}&\mathbf{m}&\mathbf{t}&\mathbf{l}\\ M&M&T&L\\ \mu&\nu&\tau&\lambda\end{array}\right)^{[\mathrm{R}^{\mathrm{I}}_{MM}\mathrm{R}^{\mathrm{I}}_{TL}]} &:= \left(\begin{array}{cc|cc}\mathbf{m}&\mathbf{m}&\mathbf{t}&\mathbf{l}\\ M&M&T&L\\ \mu&\nu&\tau&\lambda\end{array}\right)^{[\mathrm{R}^{\mathrm{I}}_{TL}]}\\
&:= \frac{1}{2}\Bigg\{\sum_{\tau'=1}^{n_o(T)}\begin{pmatrix}\mathbf{t}&\mathbf{l}\\ T&L\\ \tau'&\lambda\end{pmatrix}\left(\begin{array}{cc|cc}\mathbf{m}&\mathbf{m}&\mathbf{t}&\mathbf{t}\\ M&M&T&T\\ \mu&\nu&\tau&\tau'\end{array}\right) + \sum_{\lambda'=1}^{n_o(L)}\begin{pmatrix}\mathbf{t}&\mathbf{l}\\ T&L\\ \tau&\lambda'\end{pmatrix}\left(\begin{array}{cc|cc}\mathbf{m}&\mathbf{m}&\mathbf{l}&\mathbf{l}\\ M&M&L&L\\ \mu&\nu&\lambda'&\lambda\end{array}\right)\Bigg\},
\end{aligned} \tag{3.6}
$$

$$
\begin{aligned}
\left(\begin{array}{cc|cc}\mathbf{m}&\mathbf{t}&\mathbf{m}&\mathbf{l}\\ M&T&M&L\\ \mu&\tau&\nu&\lambda\end{array}\right)^{[\mathrm{R}^{\mathrm{I}}_{MT}\mathrm{R}^{\mathrm{I}}_{NL}]} &:= \frac{1}{2}\Bigg\{\sum_{\mu'=1}^{n_o(M)}\begin{pmatrix}\mathbf{m}&\mathbf{t}\\ M&T\\ \mu'&\tau\end{pmatrix}\left(\begin{array}{cc|cc}\mathbf{m}&\mathbf{m}&\mathbf{m}&\mathbf{l}\\ M&M&M&L\\ \mu&\mu'&\nu&\lambda\end{array}\right)^{[\mathrm{R}^{\mathrm{I}}_{ML}]} + \sum_{\tau'=1}^{n_o(T)}\begin{pmatrix}\mathbf{m}&\mathbf{t}\\ M&T\\ \mu&\tau'\end{pmatrix}\left(\begin{array}{cc|cc}\mathbf{t}&\mathbf{t}&\mathbf{m}&\mathbf{l}\\ T&T&M&L\\ \tau'&\tau&\nu&\lambda\end{array}\right)^{[\mathrm{R}^{\mathrm{I}}_{ML}]}\Bigg\}\\
&:= \frac{1}{4}\Bigg\{\sum_{\mu'=1}^{n_o(M)}\begin{pmatrix}\mathbf{m}&\mathbf{t}\\ M&T\\ \mu'&\tau\end{pmatrix}\Bigg[\sum_{\nu'=1}^{n_o(M)}\begin{pmatrix}\mathbf{m}&\mathbf{l}\\ M&L\\ \nu'&\lambda\end{pmatrix}\left(\begin{array}{cc|cc}\mathbf{m}&\mathbf{m}&\mathbf{m}&\mathbf{m}\\ M&M&M&M\\ \mu&\mu'&\nu&\nu'\end{array}\right) + \sum_{\lambda'=1}^{n_o(L)}\begin{pmatrix}\mathbf{m}&\mathbf{l}\\ M&L\\ \nu&\lambda'\end{pmatrix}\left(\begin{array}{cc|cc}\mathbf{m}&\mathbf{m}&\mathbf{l}&\mathbf{l}\\ M&M&L&L\\ \mu&\mu'&\lambda'&\lambda\end{array}\right)\Bigg]\\
&\quad + \sum_{\tau'=1}^{n_o(T)}\begin{pmatrix}\mathbf{m}&\mathbf{t}\\ M&T\\ \mu&\tau'\end{pmatrix}\Bigg[\sum_{\nu'=1}^{n_o(M)}\begin{pmatrix}\mathbf{m}&\mathbf{l}\\ M&L\\ \nu'&\lambda\end{pmatrix}\left(\begin{array}{cc|cc}\mathbf{t}&\mathbf{t}&\mathbf{m}&\mathbf{m}\\ T&T&M&M\\ \tau'&\tau&\nu&\nu'\end{array}\right) + \sum_{\lambda'=1}^{n_o(L)}\begin{pmatrix}\mathbf{m}&\mathbf{l}\\ M&L\\ \nu&\lambda'\end{pmatrix}\left(\begin{array}{cc|cc}\mathbf{t}&\mathbf{t}&\mathbf{l}&\mathbf{l}\\ T&T&L&L\\ \tau'&\tau&\lambda'&\lambda\end{array}\right)\Bigg]\Bigg\}.
\end{aligned} \tag{3.7}
$$

(II) Using two two-electron approximations of Rüdenberg type, we get

$$
\begin{aligned}
\left(\begin{array}{cc|cc}\mathbf{m}&\mathbf{m}&\mathbf{t}&\mathbf{l}\\ M&M&T&L\\ \mu&\nu&\tau&\lambda\end{array}\right)^{[\mathrm{R}^{\mathrm{II}}_{MT}\mathrm{R}^{\mathrm{II}}_{ML}]} &:= \frac{1}{2}\Bigg\{\sum_{\mu'=1}^{n_o(M)}\begin{pmatrix}\mathbf{m}&\mathbf{t}\\ M&T\\ \mu'&\tau\end{pmatrix}\left(\begin{array}{cc|cc}\mathbf{m}&\mathbf{m}&\mathbf{m}&\mathbf{l}\\ M&M&M&L\\ \mu&\nu&\mu'&\lambda\end{array}\right)^{[\mathrm{R}^{\mathrm{II}}_{ML}]} + \sum_{\tau'=1}^{n_o(T)}\begin{pmatrix}\mathbf{m}&\mathbf{t}\\ M&T\\ \mu&\tau'\end{pmatrix}\left(\begin{array}{cc|cc}\mathbf{t}&\mathbf{m}&\mathbf{t}&\mathbf{l}\\ T&M&M&L\\ \tau'&\nu&\tau&\lambda\end{array}\right)^{[\mathrm{R}^{\mathrm{II}}_{ML}]}\Bigg\}\\
&:= \frac{1}{4}\Bigg\{\sum_{\mu'=1}^{n_o(M)}\begin{pmatrix}\mathbf{m}&\mathbf{t}\\ M&T\\ \mu'&\tau\end{pmatrix}\Bigg[\sum_{\nu'=1}^{n_o(M)}\begin{pmatrix}\mathbf{m}&\mathbf{l}\\ M&L\\ \nu'&\lambda\end{pmatrix}\left(\begin{array}{cc|cc}\mathbf{m}&\mathbf{m}&\mathbf{m}&\mathbf{m}\\ M&M&M&M\\ \mu&\nu&\mu'&\nu'\end{array}\right) + \sum_{\lambda'=1}^{n_o(L)}\begin{pmatrix}\mathbf{m}&\mathbf{l}\\ M&L\\ \nu&\lambda'\end{pmatrix}\left(\begin{array}{cc|cc}\mathbf{m}&\mathbf{l}&\mathbf{m}&\mathbf{l}\\ M&L&M&L\\ \mu&\lambda'&\mu'&\lambda\end{array}\right)\Bigg]\\
&\quad + \sum_{\tau'=1}^{n_o(T)}\begin{pmatrix}\mathbf{m}&\mathbf{t}\\ M&T\\ \mu&\tau'\end{pmatrix}\Bigg[\sum_{\nu'=1}^{n_o(M)}\begin{pmatrix}\mathbf{m}&\mathbf{l}\\ M&L\\ \nu'&\lambda\end{pmatrix}\left(\begin{array}{cc|cc}\mathbf{t}&\mathbf{m}&\mathbf{t}&\mathbf{m}\\ T&M&T&M\\ \tau'&\nu&\tau&\nu'\end{array}\right) + \sum_{\lambda'=1}^{n_o(L)}\begin{pmatrix}\mathbf{m}&\mathbf{l}\\ M&L\\ \nu&\lambda'\end{pmatrix}\left(\begin{array}{cc|cc}\mathbf{t}&\mathbf{l}&\mathbf{t}&\mathbf{l}\\ T&L&T&L\\ \tau'&\lambda'&\tau&\lambda\end{array}\right)\Bigg]\Bigg\},
\end{aligned} \tag{3.8}
$$

$$
\begin{aligned}
\left(\begin{array}{cc|cc}\mathbf{m}&\mathbf{t}&\mathbf{m}&\mathbf{l}\\ M&T&M&L\\ \mu&\tau&\nu&\lambda\end{array}\right)^{[\mathrm{R}^{\mathrm{II}}_{MM}\mathrm{R}^{\mathrm{II}}_{TL}]} &:= \left(\begin{array}{cc|cc}\mathbf{m}&\mathbf{t}&\mathbf{m}&\mathbf{l}\\ M&T&M&L\\ \mu&\tau&\nu&\lambda\end{array}\right)^{[\mathrm{R}^{\mathrm{II}}_{TL}]}\\
&:= \frac{1}{2}\Bigg\{\sum_{\tau'=1}^{n_o(T)}\begin{pmatrix}\mathbf{t}&\mathbf{l}\\ T&L\\ \tau'&\lambda\end{pmatrix}\left(\begin{array}{cc|cc}\mathbf{m}&\mathbf{t}&\mathbf{m}&\mathbf{t}\\ M&T&M&T\\ \mu&\tau&\nu&\tau'\end{array}\right) + \sum_{\lambda'=1}^{n_o(L)}\begin{pmatrix}\mathbf{t}&\mathbf{l}\\ T&L\\ \tau&\lambda'\end{pmatrix}\left(\begin{array}{cc|cc}\mathbf{m}&\mathbf{l}&\mathbf{m}&\mathbf{l}\\ M&L&M&L\\ \mu&\lambda'&\nu&\lambda\end{array}\right)\Bigg\}.
\end{aligned} \tag{3.9}
$$

Hence, applying Rüdenberg's approximation twice implies an oversimplification of the two-center integral $\left(\begin{array}{cc|cc} \mathbf{m} & \mathbf{m} & \mathbf{m} & \mathbf{l} \\ M & M & M & L \\ \mu & \mu' & \nu & \lambda \end{array}\right)^{[\mathrm{R}^{\mathrm{I}}_{ML}]}$ in (3.7) and of $\left(\begin{array}{cc|cc} \mathbf{m} & \mathbf{m} & \mathbf{m} & \mathbf{l} \\ M & M & M & L \\ \mu & \nu & \mu' & \lambda \end{array}\right)^{[\mathrm{R}^{\mathrm{II}}_{ML}]}$ in (3.8). Obviously, the formulations of (3.6) and (3.9) should be preferred, since they use Rüdenberg's recipe only once. While the oversimplifying "unrestricted" branch of approximation has been discussed comprehensively in [6], we now turn to the corresponding "restricted" route, which avoids such shortcomings.

3.2. "Restricted and Combined Rüdenberg" Approximations (R.R&C) for Fock-matrix Elements

The term ' "Restricted and Combined Rüdenberg" approximations (R.R&C) ' indicates, 1) that both one-electron and two-electron routes of approximation are combined in the sense outlined in [6], and 2) that in this subsection we are going to distinguish four-center and three-center interactions from one another and those of two-center or one-center type. All different types of three-center integrals occuring in (2.1) … (2.8) will be treated in such a way that oversimplifications are avoided by applying Rüdenberg's approximations only once. Furthermore, this time all one- and two-center interactions are considered to be evaluated accurately.

Distinguishing off-blockdiagonal from blockdiagonal matrix elements we define according to ([6] 2.24)

$$\underbrace{(\mathbf{F}^{\alpha}_{\mathbf{0n}})^{[\mathrm{R.R\&C}]}_{(M,\mu)(N,\nu)}}_{\mathbf{n}N\neq\mathbf{0}M} := (\mathbf{K}_{\mathbf{0n}})_{(M,\mu)(N,\nu)} + (\mathbf{F}^{A}_{\mathbf{0n}})^{[\mathrm{R.R\&C}]}_{(M,\mu)(N,\nu)} + (\mathbf{F}^{C}_{\mathbf{0n}})^{[\mathrm{R.R\&C}]}_{(M,\mu)(N,\nu)} - (\mathbf{F}^{\alpha E}_{\mathbf{0n}})^{[\mathrm{R.R\&C}]}_{(M,\mu)(N,\nu)}, \tag{3.10}$$

$$(\mathbf{F}^{\alpha}_{\mathbf{00}})^{[\mathrm{R.R\&C}]}_{(M,\mu)(M,\nu)} := (\mathbf{K}_{\mathbf{00}})_{(M,\mu)(M,\nu)} + (\mathbf{F}^{A}_{\mathbf{00}})_{(M,\mu)(M,\nu)} + (\mathbf{F}^{C}_{\mathbf{00}})^{[\mathrm{R.R\&C}]}_{(M,\mu)(M,\nu)} - (\mathbf{F}^{\alpha E}_{\mathbf{00}})^{[\mathrm{R.R\&C}]}_{(M,\mu)(M,\nu)}. \tag{3.11}$$

For the off-blockdiagonal attractive part we define according to (2.1)

$$\underbrace{(\mathbf{F}^{A}_{\mathbf{0n}})^{[\mathrm{R.R\&C}]}_{(M,\mu)(N,\nu)}}_{\mathbf{n}N\neq\mathbf{0}M} := ({}^{(0)}\mathbf{A}_{\mathbf{0n}})_{(M,\mu)(N,\nu)} + ({}^{(1)}\mathbf{A}_{\mathbf{0n}})^{[\mathrm{R}^{\mathrm{I}}]}_{(M,\mu)(N,\nu)}. \tag{3.12}$$

For the off-blockdiagonal Coulomb part we define according to (2.3)

$$\underbrace{(\mathbf{F}^{C}_{\mathbf{0n}})^{[\mathrm{R.R\&C}]}_{(M,\mu)(N,\nu)}}_{\mathbf{n}N\neq\mathbf{0}M} := ({}^{(0)}\mathbf{C}_{\mathbf{0n}})_{(M,\mu)(N,\nu)} + ({}^{(1)}\mathbf{C}_{\mathbf{0n}})^{[\mathrm{R}^{\mathrm{I}}\mathrm{R}^{\mathrm{I}}]}_{(M,\mu)(N,\nu)} + ({}^{(2)}\mathbf{C}_{\mathbf{0n}})^{[\mathrm{R}^{\mathrm{I}}]}_{(M,\mu)(N,\nu)}$$
$$+\,2({}^{(3)}\mathbf{C}_{\mathbf{0n}})^{[\mathrm{R}^{\mathrm{II}}]}_{(M,\mu)(N,\nu)} + 2({}^{(4)}\mathbf{C}_{\mathbf{0n}})^{[\mathrm{R}^{\mathrm{II}}]}_{(M,\mu)(N,\nu)}. \tag{3.13}$$

For the blockdiagonal Coulomb part we define according to (2.5)

$$(\mathbf{F}^{C}_{\mathbf{00}})^{[\mathrm{R.R\&C}]}_{(M,\mu)(M,\nu)} := ({}^{(0)}\mathbf{C}_{\mathbf{00}})_{(M,\mu)(M,\nu)} + ({}^{(1)}\mathbf{C}_{\mathbf{00}})^{[\mathrm{R}^{\mathrm{I}}]}_{(M,\mu)(M,\nu)} + ({}^{(2)}\mathbf{C}_{\mathbf{00}})_{(M,\mu)(M,\nu)} + 2({}^{(3)}\mathbf{C}_{\mathbf{00}})_{(M,\mu)(M,\nu)}. \tag{3.14}$$

For the off-blockdiagonal exchange part we define according to (2.6)

$$\underbrace{(\mathbf{F}^{\alpha E}_{\mathbf{0n}})^{[\mathrm{R.R\&C}]}_{(M,\mu)(N,\nu)}}_{\mathbf{n}N\neq\mathbf{0}M} := ({}^{(0)}\mathbf{E}^{\alpha}_{\mathbf{0n}})_{(M,\mu)(N,\nu)} + ({}^{(1)}\mathbf{E}^{\alpha}_{\mathbf{0n}})^{[\mathrm{R}^{\mathrm{II}}\mathrm{R}^{\mathrm{II}}]}_{(M,\mu)(N,\nu)} + ({}^{(2)}\mathbf{E}^{\alpha}_{\mathbf{0n}})^{[\mathrm{R}^{\mathrm{II}}]}_{(M,\mu)(N,\nu)} + ({}^{(3)}\mathbf{E}^{\alpha}_{\mathbf{0n}})^{[\mathrm{R}^{\mathrm{II}}]}_{(M,\mu)(N,\nu)}$$
$$+\,({}^{(4)}\mathbf{E}^{\alpha}_{\mathbf{0n}})^{[\mathrm{R}^{\mathrm{I}}]}_{(M,\mu)(N,\nu)} + ({}^{(5)}\mathbf{E}^{\alpha}_{\mathbf{0n}})^{[\mathrm{R}^{\mathrm{I}}]}_{(M,\mu)(N,\nu)} + ({}^{(6)}\mathbf{E}^{\alpha}_{\mathbf{0n}})^{[\mathrm{R}^{\mathrm{II}}]}_{(M,\mu)(N,\nu)}. \tag{3.15}$$

For the blockdiagonal exchange part we define according to (2.8)

$$\begin{aligned}(\mathbf{F}^{\alpha E}_{\mathbf{00}})^{[\mathrm{R.R\&C}]}_{(M,\mu)(M,\nu)} &:= ({}^{(0)}\mathbf{E}^{\alpha}_{\mathbf{00}})_{(M,\mu)(M,\nu)} + ({}^{(1)}\mathbf{E}^{\alpha}_{\mathbf{00}})^{[\mathrm{R^{II}}]}_{(M,\mu)(M,\nu)} + ({}^{(2)}\mathbf{E}^{\alpha}_{\mathbf{00}})_{(M,\mu)(M,\nu)} \\ &\quad + ({}^{(3)}\mathbf{E}^{\alpha}_{\mathbf{00}})_{(M,\mu)(M,\nu)} + ({}^{(5)}\mathbf{E}^{\alpha}_{\mathbf{00}})_{(M,\mu)(M,\nu)}. \end{aligned} \tag{3.16}$$

Using again the notation $\mathbf{n}N \neq \mathbf{0}M \equiv (\mathbf{n} \neq \mathbf{0}) \vee (N \neq M)$, the different quantities occuring in (3.12) … (3.16) are defined as follows [15]:

$$\underbrace{({}^{(1)}\mathbf{A}_{\mathbf{0n}})^{[\mathrm{R^I}]}_{(M,\mu)(N,\nu)}}_{\mathbf{n}N\neq\mathbf{0}M} := \frac{1}{2}\left\{ \sum_{\mu'=1}^{n_o(M)} \begin{pmatrix} \mathbf{0} & \mathbf{n} \\ M & N \\ \mu' & \nu \end{pmatrix} \underbrace{\underbrace{\sum_{\mathbf{p}=\mathbf{N}^-}^{\mathbf{N}^+} \sum_{P=1}^{N_n}}_{\mathbf{p}P\neq\mathbf{0}M,\mathbf{n}N} \begin{pmatrix} \mathbf{0} & \mathbf{p} & \mathbf{0} \\ M & P & M \\ \mu & & \mu' \end{pmatrix}}_{\stackrel{\mathrm{def}}{=}({}^{(1.1)}\mathbf{A}_{\mathbf{0n}})_{(M,\mu,\mu')(N)}} + \sum_{\nu'=1}^{n_o(N)} \begin{pmatrix} \mathbf{0} & \mathbf{n} \\ M & N \\ \mu & \nu' \end{pmatrix} \underbrace{\underbrace{\sum_{\mathbf{p}=\mathbf{N}^-}^{\mathbf{N}^+} \sum_{P=1}^{N_n}}_{\mathbf{p}P\neq\mathbf{0}M,\mathbf{n}N} \begin{pmatrix} \mathbf{n} & \mathbf{p} & \mathbf{n} \\ N & P & N \\ \nu' & & \nu \end{pmatrix}}_{\stackrel{\mathrm{def}}{=}({}^{(1.2)}\mathbf{A}_{\mathbf{0n}})_{(M)(N,\nu',\nu)}} \right\}, \tag{3.17}$$

with

$$\underbrace{({}^{(1.1)}\mathbf{A}_{\mathbf{0n}})_{(M,\mu,\mu')(N)}}_{\mathbf{n}N\neq\mathbf{0}M} = \underbrace{\sum_{\mathbf{p}=\mathbf{N}^-}^{\mathbf{N}^+} \sum_{P=1}^{N_n} \begin{pmatrix} \mathbf{0} & \mathbf{p} & \mathbf{0} \\ M & P & M \\ \mu & & \mu' \end{pmatrix}}_{=(\mathbf{F}^{A}_{\mathbf{00}})_{(M,\mu)(M,\mu')}} - \begin{pmatrix} \mathbf{0} & \mathbf{0} & \mathbf{0} \\ M & M & M \\ \mu & & \mu' \end{pmatrix} - \begin{pmatrix} \mathbf{0} & \mathbf{n} & \mathbf{0} \\ M & N & M \\ \mu & & \mu' \end{pmatrix}, \tag{3.18}$$

and

$$\underbrace{({}^{(1.2)}\mathbf{A}_{\mathbf{0n}})_{(M)(N,\nu',\nu)}}_{\mathbf{n}N\neq\mathbf{0}M} = \underbrace{\sum_{\mathbf{p}=\mathbf{N}^-}^{\mathbf{N}^+} \sum_{P=1}^{N_n} \begin{pmatrix} \mathbf{n} & \mathbf{p} & \mathbf{n} \\ N & P & N \\ \nu' & & \nu \end{pmatrix}}_{=(\mathbf{F}^{A}_{\mathbf{00}})_{(N,\nu')(N,\nu)}} - \begin{pmatrix} \mathbf{n} & \mathbf{0} & \mathbf{n} \\ N & M & N \\ \nu' & & \nu \end{pmatrix} - \begin{pmatrix} \mathbf{n} & \mathbf{n} & \mathbf{n} \\ N & N & N \\ \nu' & & \nu \end{pmatrix}. \tag{3.19}$$

Introducing the abbreviations

$$(\mathbf{Q}^{\oplus}_{\mathbf{0n}})_{(M)(N,\nu,\nu')} \stackrel{\mathrm{def}}{=} \sum_{\mu=1}^{n_o(M)} (\mathbf{P}^{\oplus}_{\mathbf{0n}})_{(M,\mu)(N,\nu)} \begin{pmatrix} \mathbf{0} & \mathbf{n} \\ M & N \\ \mu & \nu' \end{pmatrix} \tag{3.20}$$

and

$$(\mathbf{Q}^{\oplus}_{\mathbf{0n}})_{(M,\mu,\mu')(N)} \stackrel{\mathrm{def}}{=} \sum_{\nu=1}^{n_o(N)} (\mathbf{P}^{\oplus}_{\mathbf{0n}})_{(M,\mu)(N,\nu)} \begin{pmatrix} \mathbf{0} & \mathbf{n} \\ M & N \\ \mu' & \nu \end{pmatrix} \tag{3.21}$$

we define:

$$\begin{aligned}\underbrace{({}^{(1)}\mathbf{C}_{\mathbf{0n}})^{[\mathrm{R^IR^I}]}_{(M,\mu)(N,\nu)}}_{\mathbf{n}N\neq\mathbf{0}M} &:= \frac{1}{2}\left\{ \sum_{\mu'=1}^{n_o(M)} \begin{pmatrix} \mathbf{0} & \mathbf{n} \\ M & N \\ \mu' & \nu \end{pmatrix} \underbrace{\underbrace{\sum_{\mathbf{t}=\mathbf{N}^-}^{\mathbf{N}^+} \sum_{T=1}^{N_n}}_{\mathbf{t}T\neq\mathbf{0}M,\mathbf{n}N} \sum_{\tau=1}^{n_o(T)} \underbrace{\sum_{\mathbf{l}=\mathbf{N}^-}^{\mathbf{N}^+} \sum_{L=1}^{N_n}}_{\mathbf{l}L\neq\mathbf{0}M,\mathbf{n}N,\mathbf{t}T} \sum_{\lambda=1}^{n_o(L)} (\mathbf{P}^{\oplus}_{\mathbf{tl}})_{(T,\tau)(L,\lambda)} \left(\begin{array}{cc|cc} \mathbf{0} & \mathbf{0} & \mathbf{t} & \mathbf{l} \\ M & M & T & L \\ \mu & \mu' & \tau & \lambda \end{array}\right)^{[\mathrm{R}^{\mathrm{I}}_{TL}]}}_{\stackrel{\mathrm{def}}{=}({}^{(1.1)}\mathbf{C}_{\mathbf{0n}})^{[\mathrm{R^I}]}_{(M,\mu,\mu')(N)}} \right. \\ &\quad \left. + \sum_{\nu'=1}^{n_o(N)} \begin{pmatrix} \mathbf{0} & \mathbf{n} \\ M & N \\ \mu & \nu' \end{pmatrix} \underbrace{\underbrace{\sum_{\mathbf{t}=\mathbf{N}^-}^{\mathbf{N}^+} \sum_{T=1}^{N_n}}_{\mathbf{t}T\neq\mathbf{0}M,\mathbf{n}N} \sum_{\tau=1}^{n_o(T)} \underbrace{\sum_{\mathbf{l}=\mathbf{N}^-}^{\mathbf{N}^+} \sum_{L=1}^{N_n}}_{\mathbf{l}L\neq\mathbf{0}M,\mathbf{n}N,\mathbf{t}T} \sum_{\lambda=1}^{n_o(L)} (\mathbf{P}^{\oplus}_{\mathbf{tl}})_{(T,\tau)(L,\lambda)} \left(\begin{array}{cc|cc} \mathbf{n} & \mathbf{n} & \mathbf{t} & \mathbf{l} \\ N & N & T & L \\ \nu' & \nu & \tau & \lambda \end{array}\right)^{[\mathrm{R}^{\mathrm{I}}_{TL}]}}_{\stackrel{\mathrm{def}}{=}({}^{(1.2)}\mathbf{C}_{\mathbf{0n}})^{[\mathrm{R^I}]}_{(M)(N,\nu',\nu)}} \right\}, \end{aligned} \tag{3.22}$$

with

$$\underbrace{\left({}^{(1.1)}\mathbf{C}_{\mathbf{0n}}\right)^{[\mathrm{R}^{\mathrm{I}}]}_{(M,\mu,\mu')(N)}}_{\mathbf{n}N\neq\mathbf{0}M} := \underbrace{\sum_{\mathbf{t}=\mathbf{N}^-}^{\mathbf{N}^+}\sum_{T=1}^{N_n}}_{\mathbf{t}T\neq\mathbf{0}M,\mathbf{n}N}\sum_{\tau=1}^{n_o(T)}\underbrace{\sum_{\mathbf{l}=\mathbf{N}^-}^{\mathbf{N}^+}\sum_{L=1}^{N_n}}_{\mathbf{l}L\neq\mathbf{0}M,\mathbf{n}N,\mathbf{t}T}\sum_{\lambda=1}^{n_o(L)}(\mathbf{P}^{\oplus}_{\mathbf{tl}})_{(T,\tau)(L,\lambda)}\frac{1}{2}\left\{\sum_{\tau'=1}^{n_o(T)}\begin{pmatrix}\mathbf{t} & \mathbf{l}\\ T & L\\ \tau' & \lambda\end{pmatrix}\left(\begin{array}{cc|cc}\mathbf{0} & \mathbf{0} & \mathbf{t} & \mathbf{t}\\ M & M & T & T\\ \mu & \mu' & \tau & \tau'\end{array}\right)\right.$$

$$\left.+\sum_{\lambda'=1}^{n_o(L)}\begin{pmatrix}\mathbf{t} & \mathbf{l}\\ T & L\\ \tau & \lambda'\end{pmatrix}\left(\begin{array}{cc|cc}\mathbf{0} & \mathbf{0} & \mathbf{l} & \mathbf{l}\\ M & M & L & L\\ \mu & \mu' & \lambda' & \lambda\end{array}\right)\right\}$$

$$=\underbrace{\sum_{\mathbf{t}=\mathbf{N}^-}^{\mathbf{N}^+}\sum_{T=1}^{N_n}}_{\mathbf{t}T\neq\mathbf{0}M}\sum_{\tau,\tau'=1}^{n_o(T)}\left(\begin{array}{cc|cc}\mathbf{0} & \mathbf{0} & \mathbf{t} & \mathbf{t}\\ M & M & T & T\\ \mu & \mu' & \tau & \tau'\end{array}\right)\Big\{((\mathbf{P}^{\oplus}\mathbf{S})_{\mathbf{00}})_{(T,\tau)(T,\tau')}-(\mathbf{P}^{\oplus}_{\mathbf{00}})_{(T,\tau)(T,\tau')}-(\mathbf{Q}^{\oplus}_{\mathbf{0t}})_{(N)(T,\tau,\tau')}-(\mathbf{Q}^{\oplus}_{\mathbf{0t}})_{(M)(T,\tau,\tau')}\Big\}$$

$$-\sum_{\tau,\tau'=1}^{n_o(N)}\left(\begin{array}{cc|cc}\mathbf{0} & \mathbf{0} & \mathbf{n} & \mathbf{n}\\ M & M & N & N\\ \mu & \mu' & \tau & \tau'\end{array}\right)\Big\{((\mathbf{P}^{\oplus}\mathbf{S})_{\mathbf{00}})_{(N,\tau)(N,\tau')}-2(\mathbf{P}^{\oplus}_{\mathbf{00}})_{(N,\tau)(N,\tau')}-(\mathbf{Q}^{\oplus}_{\mathbf{0n}})_{(M)(N,\tau,\tau')}\Big\}$$

$$-\sum_{\tau,\tau'=1}^{n_o(M)}\left(\begin{array}{cc|cc}\mathbf{0} & \mathbf{0} & \mathbf{0} & \mathbf{0}\\ M & M & M & M\\ \mu & \mu' & \tau & \tau'\end{array}\right)\Big\{(\mathbf{Q}^{\oplus}_{\mathbf{00}})_{(N)(M,\tau,\tau')}-(\mathbf{Q}^{\oplus}_{\mathbf{0n}})_{(M,\tau,\tau')(N)}\Big\}, \tag{3.23}$$

and

$$\underbrace{\left({}^{(1.2)}\mathbf{C}_{\mathbf{0n}}\right)^{[\mathrm{R}^{\mathrm{I}}]}_{(M)(N,\nu',\nu)}}_{\mathbf{n}N\neq\mathbf{0}M} :=$$

$$\underbrace{\sum_{\mathbf{t}=\mathbf{N}^-}^{\mathbf{N}^+}\sum_{T=1}^{N_n}}_{\mathbf{t}T\neq\mathbf{0}M,\mathbf{n}N}\sum_{\tau=1}^{n_o(T)}\underbrace{\sum_{\mathbf{l}=\mathbf{N}^-}^{\mathbf{N}^+}\sum_{L=1}^{N_n}}_{\mathbf{l}L\neq\mathbf{0}M,\mathbf{n}N,\mathbf{t}T}\sum_{\lambda=1}^{n_o(L)}(\mathbf{P}^{\oplus}_{\mathbf{tl}})_{(T,\tau)(L,\lambda)}\times\frac{1}{2}\left\{\sum_{\tau'=1}^{n_o(T)}\begin{pmatrix}\mathbf{t} & \mathbf{l}\\ T & L\\ \tau' & \lambda\end{pmatrix}\left(\begin{array}{cc|cc}\mathbf{n} & \mathbf{n} & \mathbf{t} & \mathbf{t}\\ N & N & T & T\\ \nu' & \nu & \tau & \tau'\end{array}\right)+\sum_{\lambda'=1}^{n_o(L)}\begin{pmatrix}\mathbf{t} & \mathbf{l}\\ T & L\\ \tau & \lambda'\end{pmatrix}\left(\begin{array}{cc|cc}\mathbf{n} & \mathbf{n} & \mathbf{l} & \mathbf{l}\\ N & N & L & L\\ \nu' & \nu & \lambda' & \lambda\end{array}\right)\right\}$$

$$=\underbrace{\sum_{\mathbf{t}=\mathbf{N}^-}^{\mathbf{N}^+}\sum_{T=1}^{N_n}}_{\mathbf{t}T\neq\mathbf{0}N}\sum_{\tau,\tau'=1}^{n_o(T)}\left(\begin{array}{cc|cc}\mathbf{0} & \mathbf{0} & \mathbf{t} & \mathbf{t}\\ N & N & T & T\\ \nu' & \nu & \tau & \tau'\end{array}\right)\Big\{((\mathbf{P}^{\oplus}\mathbf{S})_{\mathbf{00}})_{(T,\tau)(T,\tau')}-(\mathbf{P}^{\oplus}_{\mathbf{00}})_{(T,\tau)(T,\tau')}-(\mathbf{Q}^{\oplus}_{\mathbf{0t}})_{(N)(T,\tau,\tau')}-(\mathbf{Q}^{\oplus}_{\mathbf{0t}})_{(M)(T,\tau,\tau')}\Big\}$$

$$-\sum_{\tau,\tau'=1}^{n_o(M)}\left(\begin{array}{cc|cc}\mathbf{n} & \mathbf{n} & \mathbf{0} & \mathbf{0}\\ N & N & M & M\\ \nu' & \nu & \tau & \tau'\end{array}\right)\Big\{((\mathbf{P}^{\oplus}\mathbf{S})_{\mathbf{00}})_{(M,\tau)(M,\tau')}-2(\mathbf{P}^{\oplus}_{\mathbf{00}})_{(M,\tau)(M,\tau')}-(\mathbf{Q}^{\oplus}_{\mathbf{0n}})_{(M,\tau,\tau')(N)}\Big\}$$

$$-\sum_{\tau,\tau'=1}^{n_o(N)}\left(\begin{array}{cc|cc}\mathbf{0} & \mathbf{0} & \mathbf{0} & \mathbf{0}\\ N & N & N & N\\ \nu' & \nu & \tau & \tau'\end{array}\right)\Big\{(\mathbf{Q}^{\oplus}_{\mathbf{00}})_{(M)(N,\tau,\tau')}-(\mathbf{Q}^{\oplus}_{\mathbf{0n}})_{(M)(N,\tau,\tau')}\Big\}. \tag{3.24}$$

$$\left({}^{(1)}\mathbf{C}_{\mathbf{00}}\right)^{[\mathrm{R}^{\mathrm{I}}]}_{(M,\mu)(M,\nu)} :=$$

$$\underbrace{\sum_{\mathbf{t}=\mathbf{N}^-}^{\mathbf{N}^+}\sum_{T=1}^{N_n}}_{\mathbf{t}T\neq\mathbf{0}M}\sum_{\tau=1}^{n_o(T)}\underbrace{\sum_{\mathbf{l}=\mathbf{N}^-}^{\mathbf{N}^+}\sum_{L=1}^{N_n}}_{\mathbf{l}L\neq\mathbf{0}M,\mathbf{t}T}\sum_{\lambda=1}^{n_o(L)}(\mathbf{P}^{\oplus}_{\mathbf{tl}})_{(T,\tau)(L,\lambda)}\times\frac{1}{2}\left\{\sum_{\tau'=1}^{n_o(T)}\begin{pmatrix}\mathbf{t} & \mathbf{l}\\ T & L\\ \tau' & \lambda\end{pmatrix}\left(\begin{array}{cc|cc}\mathbf{0} & \mathbf{0} & \mathbf{t} & \mathbf{t}\\ M & M & T & T\\ \mu & \nu & \tau & \tau'\end{array}\right)+\sum_{\lambda'=1}^{n_o(L)}\begin{pmatrix}\mathbf{t} & \mathbf{l}\\ T & L\\ \tau & \lambda'\end{pmatrix}\left(\begin{array}{cc|cc}\mathbf{0} & \mathbf{0} & \mathbf{l} & \mathbf{l}\\ M & M & L & L\\ \mu & \nu & \lambda' & \lambda\end{array}\right)\right\}$$

$$=\underbrace{\sum_{\mathbf{t}=\mathbf{N}^-}^{\mathbf{N}^+}\sum_{T=1}^{N_n}}_{\mathbf{t}T\neq\mathbf{0}M}\sum_{\tau=1}^{n_o(T)}\sum_{\tau'=1}^{n_o(T)}\left(\begin{array}{cc|cc}\mathbf{0} & \mathbf{0} & \mathbf{t} & \mathbf{t}\\ M & M & T & T\\ \mu & \nu & \tau & \tau'\end{array}\right)\Big\{((\mathbf{P}^{\oplus}\mathbf{S})_{\mathbf{00}})_{(T,\tau)(T,\tau')}-(\mathbf{P}^{\oplus}_{\mathbf{00}})_{(T,\tau)(T,\tau')}-(\mathbf{Q}^{\oplus}_{\mathbf{0t}})_{(M)(T,\tau,\tau')}\Big\}. \tag{3.25}$$

$$\underbrace{\left({}^{(2)}\mathbf{C}_{\mathbf{0n}}\right)^{[\mathrm{R^I}]}_{(M,\mu)(N,\nu)}}_{\mathbf{n}N\neq\mathbf{0}M} := \frac{1}{2}\Bigg\{ \sum_{\mu'=1}^{n_o(M)} \begin{pmatrix} \mathbf{0} & \mathbf{n} \\ M & N \\ \mu' & \nu \end{pmatrix} \underbrace{\underbrace{\sum_{\mathbf{t}=\mathbf{N}^-}^{\mathbf{N}^+} \sum_{T=1}^{N_n}}_{\mathbf{t}T\neq\mathbf{0}M,\mathbf{n}N} \sum_{\tau,\lambda=1}^{n_o(T)} (\mathbf{P}^{\oplus}_{\mathbf{00}})_{(T,\tau)(T,\lambda)} \left(\begin{array}{cc|cc} \mathbf{0}&\mathbf{0}&\mathbf{t}&\mathbf{t}\\ M&M&T&T\\ \mu&\mu'&\tau&\lambda\end{array}\right)}_{\stackrel{\mathrm{def}}{=}\left({}^{(2.1)}\mathbf{C}_{\mathbf{0n}}\right)_{(M,\mu,\mu')(N)}}$$

$$+ \sum_{\nu'=1}^{n_o(N)} \begin{pmatrix} \mathbf{0} & \mathbf{n} \\ M & N \\ \mu & \nu' \end{pmatrix} \underbrace{\underbrace{\sum_{\mathbf{t}=\mathbf{N}^-}^{\mathbf{N}^+} \sum_{T=1}^{N_n}}_{\mathbf{t}T\neq\mathbf{0}M,\mathbf{n}N} \sum_{\tau,\lambda=1}^{n_o(T)} (\mathbf{P}^{\oplus}_{\mathbf{00}})_{(T,\tau)(T,\lambda)} \left(\begin{array}{cc|cc} \mathbf{n}&\mathbf{n}&\mathbf{t}&\mathbf{t}\\ N&N&T&T\\ \nu'&\nu&\tau&\lambda\end{array}\right)}_{\stackrel{\mathrm{def}}{=}\left({}^{(2.2)}\mathbf{C}_{\mathbf{0n}}\right)_{(M)(N,\nu',\nu)}} \Bigg\} \tag{3.26}$$

with

$$\underbrace{\left({}^{(2.1)}\mathbf{C}_{\mathbf{0n}}\right)_{(M,\mu,\mu')(N)}}_{\mathbf{n}N\neq\mathbf{0}M} = \underbrace{\sum_{\mathbf{t}=\mathbf{N}^-}^{\mathbf{N}^+} \sum_{T=1}^{N_n}}_{\mathbf{t}T\neq\mathbf{0}M} \sum_{\tau,\lambda=1}^{n_o(T)} (\mathbf{P}^{\oplus}_{\mathbf{00}})_{(T,\tau)(T,\lambda)} \left(\begin{array}{cc|cc} \mathbf{0}&\mathbf{0}&\mathbf{t}&\mathbf{t}\\ M&M&T&T\\ \mu&\mu'&\tau&\lambda\end{array}\right) - \sum_{\tau,\lambda=1}^{n_o(N)} (\mathbf{P}^{\oplus}_{\mathbf{00}})_{(N,\tau)(N,\lambda)} \left(\begin{array}{cc|cc} \mathbf{0}&\mathbf{0}&\mathbf{n}&\mathbf{n}\\ M&M&N&N\\ \mu&\mu'&\tau&\lambda\end{array}\right) \tag{3.27}$$

and

$$\underbrace{\left({}^{(2.2)}\mathbf{C}_{\mathbf{0n}}\right)_{(M)(N,\nu',\nu)}}_{\mathbf{n}N\neq\mathbf{0}M} = \underbrace{\sum_{\mathbf{t}=\mathbf{N}^-}^{\mathbf{N}^+} \sum_{T=1}^{N_n}}_{\mathbf{t}T\neq\mathbf{0}N} \sum_{\tau,\lambda=1}^{n_o(T)} (\mathbf{P}^{\oplus}_{\mathbf{00}})_{(T,\tau)(T,\lambda)} \left(\begin{array}{cc|cc} \mathbf{0}&\mathbf{0}&\mathbf{t}&\mathbf{t}\\ N&N&T&T\\ \nu'&\nu&\tau&\lambda\end{array}\right) - \sum_{\tau,\lambda=1}^{n_o(M)} (\mathbf{P}^{\oplus}_{\mathbf{00}})_{(M,\tau)(M,\lambda)} \left(\begin{array}{cc|cc} \mathbf{n}&\mathbf{n}&\mathbf{0}&\mathbf{0}\\ N&N&M&M\\ \nu'&\nu&\tau&\lambda\end{array}\right). \tag{3.28}$$

$$\underbrace{\left({}^{(3)}\mathbf{C}_{\mathbf{0n}}\right)^{[\mathrm{R^{II}}]}_{(M,\mu)(N,\nu)}}_{\mathbf{n}N\neq\mathbf{0}M} := \frac{1}{2}\Bigg\{ \underbrace{\underbrace{\sum_{\mathbf{t}=\mathbf{N}^-}^{\mathbf{N}^+} \sum_{T=1}^{N_n}}_{\mathbf{t}T\neq\mathbf{0}M,\mathbf{n}N} \sum_{\tau=1}^{n_o(T)} \sum_{\lambda=1}^{n_o(M)} (\mathbf{P}^{\oplus}_{\mathbf{0t}})_{(M,\lambda)(T,\tau)} \sum_{\nu'=1}^{n_o(N)} \begin{pmatrix} \mathbf{n} & \mathbf{t} \\ N & T \\ \nu' & \tau \end{pmatrix} \left(\begin{array}{cc|cc} \mathbf{0}&\mathbf{n}&\mathbf{n}&\mathbf{0}\\ M&N&N&M\\ \mu&\nu&\nu'&\lambda\end{array}\right)}_{\stackrel{\mathrm{def}}{=}\left({}^{(3.1)}\mathbf{C}_{\mathbf{0n}}\right)_{(M,\mu)(N,\nu)}}$$

$$+ \underbrace{\underbrace{\sum_{\mathbf{t}=\mathbf{N}^-}^{\mathbf{N}^+} \sum_{T=1}^{N_n}}_{\mathbf{t}T\neq\mathbf{0}M,\mathbf{n}N} \sum_{\tau=1}^{n_o(T)} \sum_{\lambda=1}^{n_o(M)} (\mathbf{P}^{\oplus}_{\mathbf{0t}})_{(M,\lambda)(T,\tau)} \sum_{\tau'=1}^{n_o(T)} \begin{pmatrix} \mathbf{n} & \mathbf{t} \\ N & T \\ \nu & \tau' \end{pmatrix} \left(\begin{array}{cc|cc} \mathbf{0}&\mathbf{t}&\mathbf{t}&\mathbf{0}\\ M&T&T&M\\ \mu&\tau'&\tau&\lambda\end{array}\right)}_{\stackrel{\mathrm{def}}{=}\left({}^{(3.2)}\mathbf{C}_{\mathbf{0n}}\right)_{(M,\mu)(N,\nu)}} \Bigg\} \tag{3.29}$$

with

$$\underbrace{\left({}^{(3.1)}\mathbf{C}_{\mathbf{0n}}\right)_{(M,\mu)(N,\nu)}}_{\mathbf{n}N\neq\mathbf{0}M} = \sum_{\mathbf{t}=\mathbf{N}^-}^{\mathbf{N}^+} \sum_{T=1}^{N_n} \sum_{\tau=1}^{n_o(T)} \sum_{\lambda=1}^{n_o(M)} (\mathbf{P}^{\oplus}_{\mathbf{0t}})_{(M,\lambda)(T,\tau)} \sum_{\nu'=1}^{n_o(N)} \begin{pmatrix} \mathbf{0} & \mathbf{t} \\ N & T \\ \nu' & \tau \end{pmatrix} \left(\begin{array}{cc|cc} \mathbf{0}&\mathbf{n}&\mathbf{n}&\mathbf{0}\\ M&N&N&M\\ \mu&\nu&\nu'&\lambda\end{array}\right)$$

$$- \sum_{\tau=1}^{n_o(N)} \sum_{\lambda=1}^{n_o(M)} (\mathbf{P}^{\oplus}_{\mathbf{0n}})_{(M,\lambda)(N,\tau)} \left(\begin{array}{cc|cc} \mathbf{0}&\mathbf{n}&\mathbf{n}&\mathbf{0}\\ M&N&N&M\\ \mu&\nu&\tau&\lambda\end{array}\right)$$

$$- \sum_{\tau=1}^{n_o(M)} \sum_{\lambda=1}^{n_o(M)} (\mathbf{P}^{\oplus}_{\mathbf{00}})_{(M,\lambda)(M,\tau)} \sum_{\nu'=1}^{n_o(N)} \begin{pmatrix} \mathbf{0} & \mathbf{n} \\ M & N \\ \tau & \nu' \end{pmatrix} \left(\begin{array}{cc|cc} \mathbf{0}&\mathbf{n}&\mathbf{n}&\mathbf{0}\\ M&N&N&M\\ \mu&\nu&\nu'&\lambda\end{array}\right) \tag{3.30}$$

and

$$\begin{aligned}\underbrace{\left({}^{(3.2)}\mathbf{C}_{\mathbf{0n}}\right)_{(M,\mu)(N,\nu)}}_{\mathbf{n}N\neq\mathbf{0}M} &= \underbrace{\sum_{\mathbf{t}=\mathbf{N}^-}^{\mathbf{N}^+}\sum_{T=1}^{N_n}}_{\mathbf{t}T\neq\mathbf{0}M}\sum_{\tau=1}^{n_o(T)}\sum_{\lambda=1}^{n_o(M)}(\mathbf{P}^{\oplus}_{\mathbf{0t}})_{(M,\lambda)(T,\tau)}\sum_{\tau'=1}^{n_o(T)}\begin{pmatrix}\mathbf{0}&\mathbf{t}\\N&T\\\nu&\tau'\end{pmatrix}\left(\begin{array}{cc|cc}\mathbf{0}&\mathbf{t}&\mathbf{t}&\mathbf{0}\\M&T&T&M\\\mu&\tau'&\tau&\lambda\end{array}\right)\\
&-\sum_{\tau=1}^{n_o(N)}\sum_{\lambda=1}^{n_o(M)}(\mathbf{P}^{\oplus}_{\mathbf{0n}})_{(M,\lambda)(N,\tau)}\left(\begin{array}{cc|cc}\mathbf{0}&\mathbf{n}&\mathbf{n}&\mathbf{0}\\M&N&N&M\\\mu&\nu&\tau&\lambda\end{array}\right)\\
&-\sum_{\tau=1}^{n_o(M)}\sum_{\lambda=1}^{n_o(M)}(\mathbf{P}^{\oplus}_{\mathbf{00}})_{(M,\lambda)(M,\tau)}\sum_{\tau'=1}^{n_o(M)}\left\{\begin{pmatrix}\mathbf{0}&\mathbf{n}\\M&N\\\tau'&\nu\end{pmatrix}-\begin{pmatrix}\mathbf{0}&\mathbf{0}\\M&N\\\tau'&\nu\end{pmatrix}\right\}\left(\begin{array}{cc|cc}\mathbf{0}&\mathbf{0}&\mathbf{0}&\mathbf{0}\\M&M&M&M\\\mu&\tau'&\tau&\lambda\end{array}\right).\end{aligned}\tag{3.31}$$

$$\begin{aligned}\underbrace{\left({}^{(4)}\mathbf{C}_{\mathbf{0n}}\right)^{[\mathrm{R^{II}}]}_{(M,\mu)(N,\nu)}}_{\mathbf{n}N\neq\mathbf{0}M} &:= \frac{1}{2}\Bigg\{\underbrace{\underbrace{\sum_{\mathbf{t}=\mathbf{N}^-}^{\mathbf{N}^+}\sum_{T=1}^{N_n}}_{\mathbf{t}T\neq\mathbf{0}M,\mathbf{n}N}\sum_{\tau=1}^{n_o(T)}\sum_{\lambda=1}^{n_o(N)}(\mathbf{P}^{\oplus}_{\mathbf{tn}})_{(T,\tau)(N,\lambda)}\sum_{\mu'=1}^{n_o(M)}\begin{pmatrix}\mathbf{0}&\mathbf{t}\\M&T\\\mu'&\tau\end{pmatrix}\left(\begin{array}{cc|cc}\mathbf{0}&\mathbf{n}&\mathbf{0}&\mathbf{n}\\M&N&M&N\\\mu&\nu&\mu'&\lambda\end{array}\right)}_{\stackrel{\mathrm{def}}{=}\left({}^{(4.1)}\mathbf{C}_{\mathbf{0n}}\right)_{(M,\mu)(N,\nu)}}\\
&\quad+\underbrace{\underbrace{\sum_{\mathbf{t}=\mathbf{N}^-}^{\mathbf{N}^+}\sum_{T=1}^{N_n}}_{\mathbf{t}T\neq\mathbf{0}M,\mathbf{n}N}\sum_{\tau=1}^{n_o(T)}\sum_{\lambda=1}^{n_o(N)}(\mathbf{P}^{\oplus}_{\mathbf{tn}})_{(T,\tau)(N,\lambda)}\sum_{\tau'=1}^{n_o(T)}\begin{pmatrix}\mathbf{0}&\mathbf{t}\\M&T\\\mu&\tau'\end{pmatrix}\left(\begin{array}{cc|cc}\mathbf{t}&\mathbf{n}&\mathbf{t}&\mathbf{n}\\T&N&T&N\\\tau'&\nu&\tau&\lambda\end{array}\right)}_{\stackrel{\mathrm{def}}{=}\left({}^{(4.2)}\mathbf{C}_{\mathbf{0n}}\right)_{(M,\mu)(N,\nu)}}\Bigg\}\end{aligned}\tag{3.32}$$

with

$$\begin{aligned}\underbrace{\left({}^{(4.1)}\mathbf{C}_{\mathbf{0n}}\right)_{(M,\mu)(N,\nu)}}_{\mathbf{n}N\neq\mathbf{0}M} &= \sum_{\mathbf{t}=\mathbf{N}^-}^{\mathbf{N}^+}\sum_{T=1}^{N_n}\sum_{\tau=1}^{n_o(T)}\sum_{\lambda=1}^{n_o(N)}(\mathbf{P}^{\oplus}_{\mathbf{0t}})_{(N,\lambda)(T,\tau)}\sum_{\mu'=1}^{n_o(M)}\begin{pmatrix}\mathbf{0}&\mathbf{t}\\M&T\\\mu'&\tau\end{pmatrix}\left(\begin{array}{cc|cc}\mathbf{0}&\mathbf{n}&\mathbf{0}&\mathbf{n}\\M&N&M&N\\\mu&\nu&\mu'&\lambda\end{array}\right)\\
&-\sum_{\tau=1}^{n_o(N)}\sum_{\lambda=1}^{n_o(N)}(\mathbf{P}^{\oplus}_{\mathbf{00}})_{(N,\tau)(N,\lambda)}\sum_{\mu'=1}^{n_o(M)}\begin{pmatrix}\mathbf{0}&\mathbf{n}\\M&N\\\mu'&\tau\end{pmatrix}\left(\begin{array}{cc|cc}\mathbf{0}&\mathbf{n}&\mathbf{0}&\mathbf{n}\\M&N&M&N\\\mu&\nu&\mu'&\lambda\end{array}\right)\\
&-\sum_{\tau=1}^{n_o(M)}\sum_{\lambda=1}^{n_o(N)}(\mathbf{P}^{\oplus}_{\mathbf{0n}})_{(M,\tau)(N,\lambda)}\left(\begin{array}{cc|cc}\mathbf{0}&\mathbf{n}&\mathbf{0}&\mathbf{n}\\M&N&M&N\\\mu&\nu&\tau&\lambda\end{array}\right)\end{aligned}\tag{3.33}$$

and

$$\begin{aligned}\underbrace{\left({}^{(4.2)}\mathbf{C}_{\mathbf{0n}}\right)_{(M,\mu)(N,\nu)}}_{\mathbf{n}N\neq\mathbf{0}M} &= \underbrace{\sum_{\mathbf{t}=\mathbf{N}^-}^{\mathbf{N}^+}\sum_{T=1}^{N_n}}_{\mathbf{t}T\neq\mathbf{0}N}\sum_{\tau=1}^{n_o(T)}\sum_{\lambda=1}^{n_o(N)}(\mathbf{P}^{\oplus}_{\mathbf{0t}})_{(N,\lambda)(T,\tau)}\sum_{\tau'=1}^{n_o(T)}\begin{pmatrix}\mathbf{0}&\mathbf{t}\\M&T\\\mu&\tau'\end{pmatrix}\left(\begin{array}{cc|cc}\mathbf{t}&\mathbf{0}&\mathbf{t}&\mathbf{0}\\T&N&T&N\\\tau'&\nu&\tau&\lambda\end{array}\right)\\
&-\sum_{\tau=1}^{n_o(N)}\sum_{\lambda=1}^{n_o(N)}(\mathbf{P}^{\oplus}_{\mathbf{00}})_{(N,\tau)(N,\lambda)}\sum_{\tau'=1}^{n_o(N)}\left\{\begin{pmatrix}\mathbf{0}&\mathbf{n}\\M&N\\\mu&\tau'\end{pmatrix}-\begin{pmatrix}\mathbf{0}&\mathbf{0}\\M&N\\\mu&\tau'\end{pmatrix}\right\}\left(\begin{array}{cc|cc}\mathbf{0}&\mathbf{0}&\mathbf{0}&\mathbf{0}\\N&N&N&N\\\tau'&\nu&\tau&\lambda\end{array}\right)\\
&-\sum_{\tau=1}^{n_o(M)}\sum_{\lambda=1}^{n_o(N)}(\mathbf{P}^{\oplus}_{\mathbf{0n}})_{(M,\tau)(N,\lambda)}\left(\begin{array}{cc|cc}\mathbf{0}&\mathbf{n}&\mathbf{0}&\mathbf{n}\\M&N&M&N\\\mu&\nu&\tau&\lambda\end{array}\right).\end{aligned}\tag{3.34}$$

Introducing the abbreviations

$$(\mathbf{Q}^{\alpha}_{\mathbf{0n}})_{(M)(N,\nu,\nu')} \stackrel{\text{def}}{=} \sum_{\mu=1}^{n_o(M)} (\mathbf{P}^{\alpha}_{\mathbf{0n}})_{(M,\mu)(N,\nu)} \begin{pmatrix} \mathbf{0} & \mathbf{n} \\ M & N \\ \mu & \nu' \end{pmatrix} \tag{3.35}$$

and

$$(\mathbf{Q}^{\alpha}_{\mathbf{0n}})_{(M,\mu,\mu')(N)} \stackrel{\text{def}}{=} \sum_{\nu=1}^{n_o(N)} (\mathbf{P}^{\alpha}_{\mathbf{0n}})_{(M,\mu)(N,\nu)} \begin{pmatrix} \mathbf{0} & \mathbf{n} \\ M & N \\ \mu' & \nu \end{pmatrix} \tag{3.36}$$

we define:

$$\begin{aligned}
\underbrace{({}^{(1)}\mathbf{E}^{\alpha}_{\mathbf{0n}})^{[\mathrm{R^{II}R^{II}}]}_{(M,\mu)(N,\nu)}}_{\mathbf{n}N\neq\mathbf{0}M} := \frac{1}{2}\Bigg\{ & \sum_{\mu'=1}^{n_o(M)} \begin{pmatrix} \mathbf{0} & \mathbf{n} \\ M & N \\ \mu' & \nu \end{pmatrix} \underbrace{\underbrace{\sum_{\mathbf{t}=\mathbf{N}^-}^{\mathbf{N}^+}\sum_{T=1}^{N_n}}_{\mathbf{t}T\neq\mathbf{0}M,\mathbf{n}N} \sum_{\tau=1}^{n_o(T)} \underbrace{\sum_{\mathbf{l}=\mathbf{N}^-}^{\mathbf{N}^+}\sum_{L=1}^{N_n}}_{\mathbf{l}L\neq\mathbf{0}M,\mathbf{n}N,\mathbf{t}T} \sum_{\lambda=1}^{n_o(L)} (\mathbf{P}^{\alpha}_{\mathbf{tl}})_{(T,\tau)(L,\lambda)} \left(\begin{array}{cc|cc} \mathbf{0} & \mathbf{t} & \mathbf{0} & \mathbf{l} \\ M & T & M & L \\ \mu & \tau & \mu' & \lambda \end{array}\right)^{[\mathrm{R}^{\mathrm{II}}_{TL}]}}_{\stackrel{\text{def}}{=}({}^{(1.1)}\mathbf{E}^{\alpha}_{\mathbf{0n}})^{[\ rmR^{\mathrm{II}}]}_{(M,\mu,\mu')(N)}} \\
& + \sum_{\nu'=1}^{n_o(N)} \begin{pmatrix} \mathbf{0} & \mathbf{N} \\ M & N \\ \mu & \nu' \end{pmatrix} \underbrace{\underbrace{\sum_{\mathbf{t}=\mathbf{N}^-}^{\mathbf{N}^+}\sum_{T=1}^{N_n}}_{\mathbf{t}T\neq\mathbf{0}M,\mathbf{n}N} \sum_{\tau=1}^{n_o(T)} \underbrace{\sum_{\mathbf{l}=\mathbf{N}^-}^{\mathbf{N}^+}\sum_{L=1}^{N_n}}_{\mathbf{l}L\neq\mathbf{0}M,\mathbf{n}N,\mathbf{t}T} \sum_{\lambda=1}^{n_o(L)} (\mathbf{P}^{\alpha}_{\mathbf{tl}})_{(T,\tau)(L,\lambda)} \left(\begin{array}{cc|cc} \mathbf{n} & \mathbf{t} & \mathbf{n} & \mathbf{l} \\ N & T & N & L \\ \nu' & \tau & \nu & \lambda \end{array}\right)^{[\mathrm{R}^{\mathrm{II}}_{TL}]}}_{\stackrel{\text{def}}{=}({}^{(1.2)}\mathbf{E}^{\alpha}_{\mathbf{0n}})^{[\mathrm{R^{II}}]}_{(M)(N,\nu',\nu)}} \Bigg\}
\end{aligned} \tag{3.37}$$

with

$$\begin{aligned}
&\underbrace{({}^{(1.1)}\mathbf{E}^{\alpha}_{\mathbf{0n}})^{[\mathrm{R^{II}}]}_{(M,\mu,\mu')(N)}}_{\mathbf{n}N\neq\mathbf{0}M} := \\
&\underbrace{\sum_{\mathbf{t}=\mathbf{N}^-}^{\mathbf{N}^+}\sum_{T=1}^{N_n}}_{\mathbf{t}T\neq\mathbf{0}M,\mathbf{n}N} \sum_{\tau=1}^{n_o(T)} \underbrace{\sum_{\mathbf{l}=\mathbf{N}^-}^{\mathbf{N}^+}\sum_{L=1}^{N_n}}_{\mathbf{l}L\neq\mathbf{0}M,\mathbf{n}N,\mathbf{t}T} \sum_{\lambda=1}^{n_o(L)} (\mathbf{P}^{\alpha}_{\mathbf{tl}})_{(T,\tau)(L,\lambda)} \times \frac{1}{2}\Bigg\{ \sum_{\tau'=1}^{n_o(T)} \begin{pmatrix} \mathbf{t} & \mathbf{l} \\ T & L \\ \tau' & \lambda \end{pmatrix} \left(\begin{array}{cc|cc} \mathbf{0} & \mathbf{t} & \mathbf{0} & \mathbf{t} \\ M & T & M & T \\ \mu & \tau & \mu' & \tau' \end{array}\right) + \sum_{\lambda'=1}^{n_o(L)} \begin{pmatrix} \mathbf{t} & \mathbf{l} \\ T & L \\ \tau & \lambda' \end{pmatrix} \left(\begin{array}{cc|cc} \mathbf{0} & \mathbf{l} & \mathbf{0} & \mathbf{l} \\ M & L & M & L \\ \mu & \lambda' & \mu' & \lambda \end{array}\right) \Bigg\} \\
&= \underbrace{\sum_{\mathbf{t}=\mathbf{N}^-}^{\mathbf{N}^+}\sum_{T=1}^{N_n}}_{\mathbf{t}T\neq\mathbf{0}M} \sum_{\tau,\tau'=1}^{n_o(T)} \left(\begin{array}{cc|cc} \mathbf{0} & \mathbf{t} & \mathbf{0} & \mathbf{t} \\ M & T & M & T \\ \mu & \tau & \mu' & \tau' \end{array}\right) \Bigg\{ \frac{1}{2}\Big[((\mathbf{P}^{\alpha}\mathbf{S})_{\mathbf{00}})_{(T,\tau)(T,\tau')} + ((\mathbf{P}^{\alpha}\mathbf{S})_{\mathbf{00}})_{(T,\tau')(T,\tau)} \Big] - (\mathbf{P}^{\alpha}_{\mathbf{00}})_{(T,\tau)(T,\tau')} \\
&\qquad - \frac{1}{2}\Big[(\mathbf{Q}^{\alpha}_{\mathbf{0t}})_{(N)(T,\tau,\tau')} + (\mathbf{Q}^{\alpha}_{\mathbf{0t}})_{(N)(T,\tau',\tau)} \Big] - \frac{1}{2}\Big[(\mathbf{Q}^{\alpha}_{\mathbf{0t}})_{(M)(T,\tau,\tau')} + (\mathbf{Q}^{\alpha}_{\mathbf{0t}})_{(M)(T,\tau',\tau)} \Big] \Bigg\} \\
&\quad - \sum_{\tau,\tau'=1}^{n_o(N)} \left(\begin{array}{cc|cc} \mathbf{0} & \mathbf{n} & \mathbf{0} & \mathbf{n} \\ M & N & M & N \\ \mu & \tau & \mu' & \tau' \end{array}\right) \Bigg\{ \frac{1}{2}\Big[((\mathbf{P}^{\alpha}\mathbf{S})_{\mathbf{00}})_{(N,\tau)(N,\tau')} + ((\mathbf{P}^{\alpha}\mathbf{S})_{\mathbf{00}})_{(N,\tau')(N,\tau)} \Big] \\
&\qquad - 2(\mathbf{P}^{\alpha}_{\mathbf{00}})_{(N,\tau)(N,\tau')} - \frac{1}{2}\Big[(\mathbf{Q}^{\alpha}_{\mathbf{0n}})_{(M)(N,\tau,\tau')} + (\mathbf{Q}^{\alpha}_{\mathbf{0n}})_{(M)(N,\tau',\tau)} \Big] \Bigg\} \\
&\quad - \sum_{\tau,\tau'=1}^{n_o(M)} \left(\begin{array}{cc|cc} \mathbf{0} & \mathbf{0} & \mathbf{0} & \mathbf{0} \\ M & M & M & M \\ \mu & \tau & \mu' & \tau' \end{array}\right) \Bigg\{ \frac{1}{2}\Big[(\mathbf{Q}^{\alpha}_{\mathbf{00}})_{(N)(M,\tau,\tau')} + (\mathbf{Q}^{\alpha}_{\mathbf{00}})_{(N)(M,\tau',\tau)} \Big] - \frac{1}{2}\Big[(\mathbf{Q}^{\alpha}_{\mathbf{0n}})_{(M,\tau,\tau')(N)} + (\mathbf{Q}^{\alpha}_{\mathbf{0n}})_{(M,\tau',\tau)(N)} \Big] \Bigg\}
\end{aligned} \tag{3.38}$$

and

$$\underbrace{\left({}^{(1.2)}\mathbf{E}^{\alpha}_{\mathbf{0n}}\right)^{[\mathrm{R^{II}}]}_{(M)(N,\nu',\nu)}}_{\mathbf{n}N\neq\mathbf{0}M}:=$$

$$\underbrace{\sum_{\mathbf{t}=\mathbf{N}^-}^{\mathbf{N}^+}\sum_{T=1}^{N_n}}_{\mathbf{t}T\neq\mathbf{0}M,\mathbf{n}N}\sum_{\tau=1}^{n_o(T)}\underbrace{\sum_{\mathbf{l}=\mathbf{N}^-}^{\mathbf{N}^+}\sum_{L=1}^{N_n}}_{\mathbf{l}L\neq\mathbf{0}M,\mathbf{n}N,\mathbf{t}T}\sum_{\lambda=1}^{n_o(L)}(\mathbf{P}^{\alpha}_{\mathbf{tl}})_{(T,\tau)(L,\lambda)}\times\frac{1}{2}\left\{\sum_{\tau'=1}^{n_o(T)}\begin{pmatrix}\mathbf{t} & \mathbf{l}\\ T & L\\ \tau' & \lambda\end{pmatrix}\left(\begin{array}{cc|cc}\mathbf{n} & \mathbf{t} & \mathbf{n} & \mathbf{t}\\ N & T & N & T\\ \nu' & \tau & \nu & \tau'\end{array}\right)+\sum_{\lambda'=1}^{n_o(L)}\begin{pmatrix}\mathbf{t} & \mathbf{l}\\ T & L\\ \tau & \lambda'\end{pmatrix}\left(\begin{array}{cc|cc}\mathbf{n} & \mathbf{l} & \mathbf{n} & \mathbf{l}\\ N & L & N & L\\ \nu' & \lambda' & \nu & \lambda\end{array}\right)\right\}$$

$$=\underbrace{\sum_{\mathbf{t}=\mathbf{N}^-}^{\mathbf{N}^+}\sum_{T=1}^{N_n}}_{\mathbf{t}T\neq\mathbf{0}N}\sum_{\tau,\tau'=1}^{n_o(T)}\left(\begin{array}{cc|cc}\mathbf{0} & \mathbf{t} & \mathbf{0} & \mathbf{t}\\ N & T & N & T\\ \nu' & \tau & \nu & \tau'\end{array}\right)\left\{\frac{1}{2}\Big[((\mathbf{P}^{\alpha}\mathbf{S})_{\mathbf{00}})_{(T,\tau)(T,\tau')}+((\mathbf{P}^{\alpha}\mathbf{S})_{\mathbf{00}})_{(T,\tau')(T,\tau)}\Big]-(\mathbf{P}^{\alpha}_{\mathbf{00}})_{(T,\tau)(T,\tau')}\right.$$

$$\left.-\frac{1}{2}\Big[(\mathbf{Q}^{\alpha}_{\mathbf{0t}})_{(N)(T,\tau,\tau')}+(\mathbf{Q}^{\alpha}_{\mathbf{0t}})_{(N)(T,\tau',\tau)}\Big]-\frac{1}{2}\Big[(\mathbf{Q}^{\alpha}_{\mathbf{0t}})_{(M)(T,\tau,\tau')}+(\mathbf{Q}^{\alpha}_{\mathbf{0t}})_{(M)(T,\tau',\tau)}\Big]\right\}$$

$$-\sum_{\tau,\tau'=1}^{n_o(M)}\left(\begin{array}{cc|cc}\mathbf{n} & \mathbf{0} & \mathbf{n} & \mathbf{0}\\ N & M & N & M\\ \nu' & \tau & \nu & \tau'\end{array}\right)\left\{\frac{1}{2}\Big[((\mathbf{P}^{\alpha}\mathbf{S})_{\mathbf{00}})_{(M,\tau)(M,\tau')}+((\mathbf{P}^{\alpha}\mathbf{S})_{\mathbf{00}})_{(M,\tau')(M,\tau)}\Big]\right.$$

$$\left.-2(\mathbf{P}^{\alpha}_{\mathbf{00}})_{(M,\tau)(M,\tau')}-\frac{1}{2}\Big[(\mathbf{Q}^{\alpha}_{\mathbf{0n}})_{(M,\tau,\tau')(N)}+(\mathbf{Q}^{\alpha}_{\mathbf{0n}})_{(M,\tau',\tau)(N)}\Big]\right\} \qquad (3.39)$$

$$-\sum_{\tau,\tau'=1}^{n_o(N)}\left(\begin{array}{cc|cc}\mathbf{0} & \mathbf{0} & \mathbf{0} & \mathbf{0}\\ N & N & N & N\\ \nu' & \tau & \nu & \tau'\end{array}\right)\left\{\frac{1}{2}\Big[(\mathbf{Q}^{\alpha}_{\mathbf{00}})_{(M)(N,\tau,\tau')}+(\mathbf{Q}^{\alpha}_{\mathbf{00}})_{(M)(N,\tau',\tau)}\Big]-\frac{1}{2}\Big[(\mathbf{Q}^{\alpha}_{\mathbf{0n}})_{(M)(N,\tau,\tau')}+(\mathbf{Q}^{\alpha}_{\mathbf{0n}})_{(M)(N,\tau',\tau)}\Big]\right\}.$$

$$\left({}^{(1)}\mathbf{E}^{\alpha}_{\mathbf{00}}\right)^{[\mathrm{R^{II}}]}_{(M,\mu)(M,\nu)}:=$$

$$\underbrace{\sum_{\mathbf{t}=\mathbf{N}^-}^{\mathbf{N}^+}\sum_{T=1}^{N_n}}_{\mathbf{t}T\neq\mathbf{0}M}\sum_{\tau=1}^{n_o(T)}\underbrace{\sum_{\mathbf{l}=\mathbf{N}^-}^{\mathbf{N}^+}\sum_{L=1}^{N_n}}_{\mathbf{l}L\neq\mathbf{0}M,\mathbf{t}T}\sum_{\lambda=1}^{n_o(L)}(\mathbf{P}^{\alpha}_{\mathbf{tl}})_{(T,\tau)(L,\lambda)}\times\frac{1}{2}\left\{\sum_{\tau'=1}^{n_o(T)}\begin{pmatrix}\mathbf{t} & \mathbf{l}\\ T & L\\ \tau' & \lambda\end{pmatrix}\left(\begin{array}{cc|cc}\mathbf{0} & \mathbf{t} & \mathbf{0} & \mathbf{t}\\ M & T & M & T\\ \mu & \tau & \nu & \tau'\end{array}\right)+\sum_{\lambda'=1}^{n_o(L)}\begin{pmatrix}\mathbf{t} & \mathbf{l}\\ T & L\\ \tau & \lambda'\end{pmatrix}\left(\begin{array}{cc|cc}\mathbf{0} & \mathbf{l} & \mathbf{0} & \mathbf{l}\\ M & L & M & L\\ \mu & \lambda' & \nu & \lambda\end{array}\right)\right\}$$

$$=\underbrace{\sum_{\mathbf{t}=\mathbf{N}^-}^{\mathbf{N}^+}\sum_{T=1}^{N_n}}_{\mathbf{t}T\neq\mathbf{0}M}\sum_{\tau=1}^{n_o(T)}\sum_{\tau'=1}^{n_o(T)}\left(\begin{array}{cc|cc}\mathbf{0} & \mathbf{t} & \mathbf{0} & \mathbf{t}\\ M & T & M & T\\ \mu & \tau & \nu & \tau'\end{array}\right)\left\{\frac{1}{2}\Big[((\mathbf{P}^{\alpha}\mathbf{S})_{\mathbf{00}})_{(T,\tau)(T,\tau')}+((\mathbf{P}^{\alpha}\mathbf{S})_{\mathbf{00}})_{(T,\tau')(T,\tau)}\Big]\right.$$

$$\left.-(\mathbf{P}^{\alpha}_{\mathbf{00}})_{(T,\tau)(T,\tau')}-\frac{1}{2}\Big[(\mathbf{Q}^{\alpha}_{\mathbf{0t}})_{(M)(T,\tau,\tau')}+(\mathbf{Q}^{\alpha}_{\mathbf{0t}})_{(M)(T,\tau',\tau)}\Big]\right\}. \qquad (3.40)$$

$$\begin{aligned}\underbrace{\left({}^{(2)}\mathbf{E}^{\alpha}_{\mathbf{0n}}\right)^{[\mathrm{R^{II}}]}_{(M,\mu)(N,\nu)}}_{\mathbf{n}N\neq\mathbf{0}M} := \frac{1}{2}\Bigg\{ & \sum_{\mu'=1}^{n_o(M)} \begin{pmatrix} \mathbf{0} & \mathbf{n} \\ M & N \\ \mu' & \nu \end{pmatrix} \underbrace{\underbrace{\sum_{\mathbf{t}=\mathbf{N}^-}^{\mathbf{N}^+}\sum_{T=1}^{N_n}}_{\mathbf{t}T\neq\mathbf{0}M,\mathbf{n}N} \sum_{\tau,\lambda=1}^{n_o(T)} (\mathbf{P}^{\alpha}_{\mathbf{00}})_{(T,\tau)(T,\lambda)} \left(\begin{array}{cc|cc} \mathbf{0} & \mathbf{t} & \mathbf{0} & \mathbf{t} \\ M & T & M & T \\ \mu & \tau & \mu' & \lambda \end{array}\right)}_{\stackrel{\text{def}}{=}\left({}^{(2.1)}\mathbf{E}^{\alpha}_{\mathbf{0n}}\right)_{(M,\mu,\mu')(N)}} \\ & + \sum_{\nu'=1}^{n_o(N)} \begin{pmatrix} \mathbf{0} & \mathbf{n} \\ M & N \\ \mu & \nu' \end{pmatrix} \underbrace{\underbrace{\sum_{\mathbf{t}=\mathbf{N}^-}^{\mathbf{N}^+}\sum_{T=1}^{N_n}}_{\mathbf{t}T\neq\mathbf{0}M,\mathbf{n}N} \sum_{\tau,\lambda=1}^{n_o(T)} (\mathbf{P}^{\alpha}_{\mathbf{00}})_{(T,\tau)(T,\lambda)} \left(\begin{array}{cc|cc} \mathbf{n} & \mathbf{t} & \mathbf{n} & \mathbf{t} \\ N & T & N & T \\ \nu' & \tau & \nu & \lambda \end{array}\right)}_{\stackrel{\text{def}}{=}\left({}^{(2.2)}\mathbf{E}^{\alpha}_{\mathbf{0n}}\right)_{(M)(N,\nu',\nu)}} \Bigg\},\end{aligned} \tag{3.41}$$

with

$$\underbrace{\left({}^{(2.1)}\mathbf{E}^{\alpha}_{\mathbf{0n}}\right)_{(M,\mu,\mu')(N)}}_{\mathbf{n}N\neq\mathbf{0}M} = \underbrace{\sum_{\mathbf{t}=\mathbf{N}^-}^{\mathbf{N}^+}\sum_{T=1}^{N_n}}_{\mathbf{t}T\neq\mathbf{0}M} \sum_{\tau,\lambda=1}^{n_o(T)} (\mathbf{P}^{\alpha}_{\mathbf{00}})_{(T,\tau)(T,\lambda)} \left(\begin{array}{cc|cc} \mathbf{0} & \mathbf{t} & \mathbf{0} & \mathbf{t} \\ M & T & M & T \\ \mu & \tau & \mu' & \lambda \end{array}\right) - \sum_{\tau,\lambda=1}^{n_o(N)} (\mathbf{P}^{\alpha}_{\mathbf{00}})_{(N,\tau)(N,\lambda)} \left(\begin{array}{cc|cc} \mathbf{0} & \mathbf{n} & \mathbf{0} & \mathbf{n} \\ M & N & M & N \\ \mu & \tau & \mu' & \lambda \end{array}\right), \tag{3.42}$$

and

$$\underbrace{\left({}^{(2.2)}\mathbf{E}^{\alpha}_{\mathbf{0n}}\right)_{(M)(N,\nu',\nu)}}_{\mathbf{n}N\neq\mathbf{0}M} = \underbrace{\sum_{\mathbf{t}=\mathbf{N}^-}^{\mathbf{N}^+}\sum_{T=1}^{N_n}}_{\mathbf{t}T\neq\mathbf{0}N} \sum_{\tau,\lambda=1}^{n_o(T)} (\mathbf{P}^{\alpha}_{\mathbf{00}})_{(T,\tau)(T,\lambda)} \left(\begin{array}{cc|cc} \mathbf{0} & \mathbf{t} & \mathbf{0} & \mathbf{t} \\ N & T & N & T \\ \nu' & \tau & \nu & \lambda \end{array}\right) - \sum_{\tau,\lambda=1}^{n_o(M)} (\mathbf{P}^{\alpha}_{\mathbf{00}})_{(M,\tau)(M,\lambda)} \left(\begin{array}{cc|cc} \mathbf{n} & \mathbf{0} & \mathbf{n} & \mathbf{0} \\ N & M & N & M \\ \nu' & \tau & \nu & \lambda \end{array}\right). \tag{3.43}$$

$$\begin{aligned}\underbrace{\left({}^{(3)}\mathbf{E}^{\alpha}_{\mathbf{0n}}\right)^{[\mathrm{R^{II}}]}_{(M,\mu)(N,\nu)}}_{\mathbf{n}N\neq\mathbf{0}M} := \frac{1}{2}\Bigg\{ & \underbrace{\underbrace{\sum_{\mathbf{t}=\mathbf{N}^-}^{\mathbf{N}^+}\sum_{T=1}^{N_n}}_{\mathbf{t}T\neq\mathbf{0}M,\mathbf{n}N} \sum_{\tau=1}^{n_o(T)}\sum_{\lambda=1}^{n_o(M)} (\mathbf{P}^{\alpha}_{\mathbf{0t}})_{(M,\lambda)(T,\tau)} \sum_{\nu'=1}^{n_o(N)} \begin{pmatrix} \mathbf{n} & \mathbf{t} \\ N & T \\ \nu' & \tau \end{pmatrix} \left(\begin{array}{cc|cc} \mathbf{0} & \mathbf{n} & \mathbf{n} & \mathbf{0} \\ M & N & N & M \\ \mu & \nu' & \nu & \lambda \end{array}\right)}_{\stackrel{\text{def}}{=}\left({}^{(3.1)}\mathbf{E}^{\alpha}_{\mathbf{0n}}\right)_{(M,\mu)(N,\nu)}} \\ & + \underbrace{\underbrace{\sum_{\mathbf{t}=\mathbf{N}^-}^{\mathbf{N}^+}\sum_{T=1}^{N_n}}_{\mathbf{t}T\neq\mathbf{0}M,\mathbf{n}N} \sum_{\tau=1}^{n_o(T)}\sum_{\lambda=1}^{n_o(M)} (\mathbf{P}^{\alpha}_{\mathbf{0t}})_{(M,\lambda)(T,\tau)} \sum_{\tau'=1}^{n_o(T)} \begin{pmatrix} \mathbf{n} & \mathbf{t} \\ N & T \\ \nu & \tau' \end{pmatrix} \left(\begin{array}{cc|cc} \mathbf{0} & \mathbf{t} & \mathbf{t} & \mathbf{0} \\ M & T & T & M \\ \mu & \tau & \tau' & \lambda \end{array}\right)}_{\stackrel{\text{def}}{=}\left({}^{(3.2)}\mathbf{E}^{\alpha}_{\mathbf{0n}}\right)_{(M,\mu)(N,\nu)}} \Bigg\}\end{aligned} \tag{3.44}$$

with

$$\begin{aligned}\underbrace{\left({}^{(3.1)}\mathbf{E}^{\alpha}_{\mathbf{0n}}\right)_{(M,\mu)(N,\nu)}}_{\mathbf{n}N\neq\mathbf{0}M} = & \sum_{\mathbf{t}=\mathbf{N}^-}^{\mathbf{N}^+}\sum_{T=1}^{N_n}\sum_{\tau=1}^{n_o(T)}\sum_{\lambda=1}^{n_o(M)} (\mathbf{P}^{\alpha}_{\mathbf{0t}})_{(M,\lambda)(T,\tau)} \sum_{\nu'=1}^{n_o(N)} \begin{pmatrix} \mathbf{0} & \mathbf{t} \\ N & T \\ \nu' & \tau \end{pmatrix} \left(\begin{array}{cc|cc} \mathbf{0} & \mathbf{n} & \mathbf{n} & \mathbf{0} \\ M & N & N & M \\ \mu & \nu' & \nu & \lambda \end{array}\right) \\ & - \sum_{\tau=1}^{n_o(N)}\sum_{\lambda=1}^{n_o(M)} (\mathbf{P}^{\alpha}_{\mathbf{0n}})_{(M,\lambda)(N,\tau)} \left(\begin{array}{cc|cc} \mathbf{0} & \mathbf{n} & \mathbf{n} & \mathbf{0} \\ M & N & N & M \\ \mu & \tau & \nu & \lambda \end{array}\right) \\ & - \sum_{\tau=1}^{n_o(M)}\sum_{\lambda=1}^{n_o(M)} (\mathbf{P}^{\alpha}_{\mathbf{00}})_{(M,\lambda)(M,\tau)} \sum_{\nu'=1}^{n_o(N)} \begin{pmatrix} \mathbf{0} & \mathbf{n} \\ M & N \\ \tau & \nu' \end{pmatrix} \left(\begin{array}{cc|cc} \mathbf{0} & \mathbf{n} & \mathbf{n} & \mathbf{0} \\ M & N & N & M \\ \mu & \nu' & \nu & \lambda \end{array}\right)\end{aligned} \tag{3.45}$$

and

$$\begin{aligned}
\underbrace{\left({}^{(3.2)}\mathbf{E}^{\alpha}_{\mathbf{0n}}\right)_{(M,\mu)(N,\nu)}}_{\mathbf{n}N\neq\mathbf{0}M} = & \underbrace{\sum_{\mathbf{t}=\mathbf{N}^-}^{\mathbf{N}^+}\sum_{T=1}^{N_n}}_{\mathbf{t}T\neq\mathbf{0}M}\sum_{\tau=1}^{n_o(T)}\sum_{\lambda=1}^{n_o(M)} (\mathbf{P}^{\alpha}_{\mathbf{0t}})_{(M,\lambda)(T,\tau)} \sum_{\tau'=1}^{n_o(T)} \begin{pmatrix}\mathbf{0}&\mathbf{t}\\ N&T\\ \nu&\tau'\end{pmatrix}\left(\begin{array}{cc|cc}\mathbf{0}&\mathbf{t}&\mathbf{t}&\mathbf{0}\\ M&T&T&M\\ \mu&\tau&\tau'&\lambda\end{array}\right)\\
& -\sum_{\tau=1}^{n_o(N)}\sum_{\lambda=1}^{n_o(M)} (\mathbf{P}^{\alpha}_{\mathbf{0n}})_{(M,\lambda)(N,\tau)} \left(\begin{array}{cc|cc}\mathbf{0}&\mathbf{n}&\mathbf{n}&\mathbf{0}\\ M&N&N&M\\ \mu&\tau&\nu&\lambda\end{array}\right)\\
& -\sum_{\tau=1}^{n_o(M)}\sum_{\lambda=1}^{n_o(M)} (\mathbf{P}^{\alpha}_{\mathbf{00}})_{(M,\lambda)(M,\tau)} \sum_{\tau'=1}^{n_o(M)} \left\{\begin{pmatrix}\mathbf{0}&\mathbf{n}\\ M&N\\ \tau'&\nu\end{pmatrix}-\begin{pmatrix}\mathbf{0}&\mathbf{0}\\ M&N\\ \tau'&\nu\end{pmatrix}\right\}\left(\begin{array}{cc|cc}\mathbf{0}&\mathbf{0}&\mathbf{0}&\mathbf{0}\\ M&M&M&M\\ \mu&\tau&\tau'&\lambda\end{array}\right).
\end{aligned} \tag{3.46}$$

$$\begin{aligned}
\underbrace{\left({}^{(4)}\mathbf{E}^{\alpha}_{\mathbf{0n}}\right)^{[\mathrm{R^I}]}_{(M,\mu)(N,\nu)}}_{\mathbf{n}N\neq\mathbf{0}M} := \frac{1}{2}\Bigg\{ & \underbrace{\underbrace{\sum_{\mathbf{t}=\mathbf{N}^-}^{\mathbf{N}^+}\sum_{T=1}^{N_n}}_{\mathbf{t}T\neq\mathbf{0}M,\mathbf{n}N}\sum_{\tau=1}^{n_o(T)}\sum_{\lambda=1}^{n_o(N)} (\mathbf{P}^{\alpha}_{\mathbf{tn}})_{(T,\tau)(N,\lambda)} \sum_{\mu'=1}^{n_o(M)} \begin{pmatrix}\mathbf{0}&\mathbf{t}\\ M&T\\ \mu'&\tau\end{pmatrix}\left(\begin{array}{cc|cc}\mathbf{0}&\mathbf{0}&\mathbf{n}&\mathbf{n}\\ M&M&N&N\\ \mu&\mu'&\nu&\lambda\end{array}\right)}_{\stackrel{\text{def}}{=}\left({}^{(4.1)}\mathbf{E}^{\alpha}_{\mathbf{0n}}\right)_{(M,\mu)(N,\nu)}}\\
& +\underbrace{\underbrace{\sum_{\mathbf{t}=\mathbf{N}^-}^{\mathbf{N}^+}\sum_{T=1}^{N_n}}_{\mathbf{t}T\neq\mathbf{0}M,\mathbf{n}N}\sum_{\tau=1}^{n_o(T)}\sum_{\lambda=1}^{n_o(N)} (\mathbf{P}^{\alpha}_{\mathbf{tn}})_{(T,\tau)(N,\lambda)} \sum_{\tau'=1}^{n_o(T)} \begin{pmatrix}\mathbf{0}&\mathbf{t}\\ M&T\\ \mu&\tau'\end{pmatrix}\left(\begin{array}{cc|cc}\mathbf{t}&\mathbf{t}&\mathbf{n}&\mathbf{n}\\ T&T&N&N\\ \tau'&\tau&\nu&\lambda\end{array}\right)}_{\stackrel{\text{def}}{=}\left({}^{(4.2)}\mathbf{E}^{\alpha}_{\mathbf{0n}}\right)_{(M,\mu)(N,\nu)}}\Bigg\}
\end{aligned} \tag{3.47}$$

with

$$\begin{aligned}
\underbrace{\left({}^{(4.1)}\mathbf{E}^{\alpha}_{\mathbf{0n}}\right)_{(M,\mu)(N,\nu)}}_{\mathbf{n}N\neq\mathbf{0}M} = & \sum_{\mathbf{t}=\mathbf{N}^-}^{\mathbf{N}^+}\sum_{T=1}^{N_n}\sum_{\tau=1}^{n_o(T)}\sum_{\lambda=1}^{n_o(N)} (\mathbf{P}^{\alpha}_{\mathbf{0t}})_{(N,\lambda)(T,\tau)} \sum_{\mu'=1}^{n_o(M)} \begin{pmatrix}\mathbf{0}&\mathbf{t}\\ M&T\\ \mu'&\tau\end{pmatrix}\left(\begin{array}{cc|cc}\mathbf{0}&\mathbf{0}&\mathbf{n}&\mathbf{n}\\ M&M&N&N\\ \mu&\mu'&\nu&\lambda\end{array}\right)\\
& -\sum_{\tau=1}^{n_o(N)}\sum_{\lambda=1}^{n_o(N)} (\mathbf{P}^{\alpha}_{\mathbf{00}})_{(N,\tau)(N,\lambda)} \sum_{\mu'=1}^{n_o(M)} \begin{pmatrix}\mathbf{0}&\mathbf{n}\\ M&N\\ \mu'&\tau\end{pmatrix}\left(\begin{array}{cc|cc}\mathbf{0}&\mathbf{0}&\mathbf{n}&\mathbf{n}\\ M&M&N&N\\ \mu&\mu'&\nu&\lambda\end{array}\right)\\
& -\sum_{\tau=1}^{n_o(M)}\sum_{\lambda=1}^{n_o(N)} (\mathbf{P}^{\alpha}_{\mathbf{0n}})_{(M,\tau)(N,\lambda)} \left(\begin{array}{cc|cc}\mathbf{0}&\mathbf{0}&\mathbf{n}&\mathbf{n}\\ M&M&N&N\\ \mu&\tau&\nu&\lambda\end{array}\right)
\end{aligned} \tag{3.48}$$

and

$$\begin{aligned}
\underbrace{\left({}^{(4.2)}\mathbf{E}^{\alpha}_{\mathbf{0n}}\right)_{(M,\mu)(N,\nu)}}_{\mathbf{n}N\neq\mathbf{0}M} = & \underbrace{\sum_{\mathbf{t}=\mathbf{N}^-}^{\mathbf{N}^+}\sum_{T=1}^{N_n}}_{\mathbf{t}T\neq\mathbf{0}N}\sum_{\tau=1}^{n_o(T)}\sum_{\lambda=1}^{n_o(N)} (\mathbf{P}^{\alpha}_{\mathbf{0t}})_{(N,\lambda)(T,\tau)} \sum_{\tau'=1}^{n_o(T)} \begin{pmatrix}\mathbf{0}&\mathbf{t}\\ M&T\\ \mu&\tau'\end{pmatrix}\left(\begin{array}{cc|cc}\mathbf{t}&\mathbf{t}&\mathbf{0}&\mathbf{0}\\ T&T&N&N\\ \tau'&\tau&\nu&\lambda\end{array}\right)\\
& -\sum_{\tau=1}^{n_o(N)}\sum_{\lambda=1}^{n_o(N)} (\mathbf{P}^{\alpha}_{\mathbf{00}})_{(N,\tau)(N,\lambda)} \sum_{\tau'=1}^{n_o(N)} \left\{\begin{pmatrix}\mathbf{0}&\mathbf{n}\\ M&N\\ \mu&\tau'\end{pmatrix}-\begin{pmatrix}\mathbf{0}&\mathbf{0}\\ M&N\\ \mu&\tau'\end{pmatrix}\right\}\left(\begin{array}{cc|cc}\mathbf{0}&\mathbf{0}&\mathbf{0}&\mathbf{0}\\ N&N&N&N\\ \tau'&\tau&\nu&\lambda\end{array}\right)\\
& -\sum_{\tau=1}^{n_o(M)}\sum_{\lambda=1}^{n_o(N)} (\mathbf{P}^{\alpha}_{\mathbf{0n}})_{(M,\tau)(N,\lambda)} \left(\begin{array}{cc|cc}\mathbf{0}&\mathbf{0}&\mathbf{n}&\mathbf{n}\\ M&M&N&N\\ \mu&\tau&\nu&\lambda\end{array}\right).
\end{aligned} \tag{3.49}$$

$$
\begin{aligned}
\underbrace{\left({}^{(5)}\mathbf{E}^{\alpha}_{\mathbf{0n}}\right)^{[\mathrm{R^{I}}]}_{(M,\mu)(N,\nu)}}_{\mathbf{n}N\neq\mathbf{0}M} := \frac{1}{2}\Bigg\{ & \underbrace{\underbrace{\sum_{\mathbf{l}=\mathbf{N}^-}^{\mathbf{N}^+}\sum_{L=1}^{N_n}}_{\mathbf{l}L\neq\mathbf{0}M,\mathbf{n}N}\sum_{\tau=1}^{n_o(M)}\sum_{\lambda=1}^{n_o(L)}\left(\mathbf{P}^{\alpha}_{\mathbf{0l}}\right)_{(M,\tau)(L,\lambda)}\sum_{\nu'=1}^{n_o(N)}\begin{pmatrix}\mathbf{n} & \mathbf{l}\\ N & L\\ \nu' & \lambda\end{pmatrix}\left(\begin{array}{cc|cc}\mathbf{0} & \mathbf{0} & \mathbf{n} & \mathbf{n}\\ M & M & N & N\\ \mu & \tau & \nu & \nu'\end{array}\right)}_{\stackrel{\text{def}}{=}\left({}^{(5.1)}\mathbf{E}^{\alpha}_{\mathbf{0n}}\right)_{(M,\mu)(N,\nu)}}\\
& + \underbrace{\underbrace{\sum_{\mathbf{l}=\mathbf{N}^-}^{\mathbf{N}^+}\sum_{L=1}^{N_n}}_{\mathbf{l}L\neq\mathbf{0}M,\mathbf{n}N}\sum_{\tau=1}^{n_o(M)}\sum_{\lambda=1}^{n_o(L)}\left(\mathbf{P}^{\alpha}_{\mathbf{0l}}\right)_{(M,\tau)(L,\lambda)}\sum_{\lambda'=1}^{n_o(L)}\begin{pmatrix}\mathbf{n} & \mathbf{l}\\ N & L\\ \nu & \lambda'\end{pmatrix}\left(\begin{array}{cc|cc}\mathbf{0} & \mathbf{0} & \mathbf{l} & \mathbf{l}\\ M & M & L & L\\ \mu & \tau & \lambda' & \lambda\end{array}\right)}_{\stackrel{\text{def}}{=}\left({}^{(5.2)}\mathbf{E}^{\alpha}_{\mathbf{0n}}\right)_{(M,\mu)(N,\nu)}}\Bigg\}
\end{aligned}
\tag{3.50}
$$

with

$$
\begin{aligned}
\underbrace{\left({}^{(5.1)}\mathbf{E}^{\alpha}_{\mathbf{0n}}\right)_{(M,\mu)(N,\nu)}}_{\mathbf{n}N\neq\mathbf{0}M} = & \sum_{\mathbf{l}=\mathbf{N}^-}^{\mathbf{N}^+}\sum_{L=1}^{N_n}\sum_{\tau=1}^{n_o(M)}\sum_{\lambda=1}^{n_o(L)}\left(\mathbf{P}^{\alpha}_{\mathbf{0l}}\right)_{(M,\tau)(L,\lambda)}\sum_{\nu'=1}^{n_o(N)}\begin{pmatrix}\mathbf{0} & \mathbf{l}\\ N & L\\ \nu' & \lambda\end{pmatrix}\left(\begin{array}{cc|cc}\mathbf{0} & \mathbf{0} & \mathbf{n} & \mathbf{n}\\ M & M & N & N\\ \mu & \tau & \nu & \nu'\end{array}\right)\\
& - \sum_{\tau=1}^{n_o(M)}\sum_{\lambda=1}^{n_o(N)}\left(\mathbf{P}^{\alpha}_{\mathbf{0n}}\right)_{(M,\tau)(N,\lambda)}\left(\begin{array}{cc|cc}\mathbf{0} & \mathbf{0} & \mathbf{n} & \mathbf{n}\\ M & M & N & N\\ \mu & \tau & \nu & \lambda\end{array}\right)\\
& - \sum_{\tau=1}^{n_o(M)}\sum_{\lambda=1}^{n_o(M)}\left(\mathbf{P}^{\alpha}_{\mathbf{00}}\right)_{(M,\tau)(M,\lambda)}\sum_{\nu'=1}^{n_o(N)}\begin{pmatrix}\mathbf{0} & \mathbf{n}\\ M & N\\ \lambda & \nu'\end{pmatrix}\left(\begin{array}{cc|cc}\mathbf{0} & \mathbf{0} & \mathbf{n} & \mathbf{n}\\ M & M & N & N\\ \mu & \tau & \nu & \nu'\end{array}\right)
\end{aligned}
\tag{3.51}
$$

and

$$
\begin{aligned}
\underbrace{\left({}^{(5.2)}\mathbf{E}^{\alpha}_{\mathbf{0n}}\right)_{(M,\mu)(N,\nu)}}_{\mathbf{n}N\neq\mathbf{0}M} = & \underbrace{\sum_{\mathbf{l}=\mathbf{N}^-}^{\mathbf{N}^+}\sum_{L=1}^{N_n}}_{\mathbf{l}L\neq\mathbf{0}M}\sum_{\tau=1}^{n_o(M)}\sum_{\lambda=1}^{n_o(L)}\left(\mathbf{P}^{\alpha}_{\mathbf{0l}}\right)_{(M,\tau)(L,\lambda)}\sum_{\lambda'=1}^{n_o(L)}\begin{pmatrix}\mathbf{0} & \mathbf{l}\\ N & L\\ \nu & \lambda'\end{pmatrix}\left(\begin{array}{cc|cc}\mathbf{0} & \mathbf{0} & \mathbf{l} & \mathbf{l}\\ M & M & L & L\\ \mu & \tau & \lambda' & \lambda\end{array}\right)\\
& - \sum_{\tau=1}^{n_o(M)}\sum_{\lambda=1}^{n_o(N)}\left(\mathbf{P}^{\alpha}_{\mathbf{0n}}\right)_{(M,\tau)(N,\lambda)}\left(\begin{array}{cc|cc}\mathbf{0} & \mathbf{0} & \mathbf{n} & \mathbf{n}\\ M & M & N & N\\ \mu & \tau & \nu & \lambda\end{array}\right)\\
& - \sum_{\tau=1}^{n_o(M)}\sum_{\lambda=1}^{n_o(M)}\left(\mathbf{P}^{\alpha}_{\mathbf{00}}\right)_{(M,\tau)(M,\lambda)}\sum_{\lambda'=1}^{n_o(M)}\left\{\begin{pmatrix}\mathbf{0} & \mathbf{n}\\ M & N\\ \lambda' & \nu\end{pmatrix} - \begin{pmatrix}\mathbf{0} & \mathbf{0}\\ M & N\\ \lambda' & \nu\end{pmatrix}\right\}\left(\begin{array}{cc|cc}\mathbf{0} & \mathbf{0} & \mathbf{0} & \mathbf{0}\\ M & M & M & M\\ \mu & \tau & \lambda' & \lambda\end{array}\right).
\end{aligned}
\tag{3.52}
$$

$$
\begin{aligned}
\underbrace{\left({}^{(6)}\mathbf{E}^{\alpha}_{\mathbf{0n}}\right)^{[\mathrm{R^{II}}]}_{(M,\mu)(N,\nu)}}_{\mathbf{n}N\neq\mathbf{0}M} := \frac{1}{2}\Bigg\{ & \underbrace{\underbrace{\sum_{\mathbf{l}=\mathbf{N}^-}^{\mathbf{N}^+}\sum_{L=1}^{N_n}}_{\mathbf{l}L\neq\mathbf{0}M,\mathbf{n}N}\sum_{\tau=1}^{n_o(N)}\sum_{\lambda=1}^{n_o(L)}\left(\mathbf{P}^{\alpha}_{\mathbf{nl}}\right)_{(N,\tau)(L,\lambda)}\sum_{\mu'=1}^{n_o(M)}\begin{pmatrix}\mathbf{0} & \mathbf{l}\\ M & L\\ \mu' & \lambda\end{pmatrix}\left(\begin{array}{cc|cc}\mathbf{0} & \mathbf{n} & \mathbf{n} & \mathbf{0}\\ M & N & N & M\\ \mu & \tau & \nu & \mu'\end{array}\right)}_{\stackrel{\text{def}}{=}\left({}^{(6.1)}\mathbf{E}^{\alpha}_{\mathbf{0n}}\right)_{(M,\mu)(N,\nu)}}\\
& + \underbrace{\underbrace{\sum_{\mathbf{l}=\mathbf{n}^-}^{\mathbf{N}^+}\sum_{L=1}^{N_n}}_{\mathbf{l}L\neq\mathbf{0}M,\mathbf{N}N}\sum_{\tau=1}^{n_o(N)}\sum_{\lambda=1}^{n_o(L)}\left(\mathbf{P}^{\alpha}_{\mathbf{nl}}\right)_{(N,\tau)(L,\lambda)}\sum_{\lambda'=1}^{n_o(L)}\begin{pmatrix}\mathbf{0} & \mathbf{l}\\ M & L\\ \mu & \lambda'\end{pmatrix}\left(\begin{array}{cc|cc}\mathbf{l} & \mathbf{n} & \mathbf{n} & \mathbf{l}\\ L & N & N & L\\ \lambda' & \tau & \nu & \lambda\end{array}\right)}_{\stackrel{\text{def}}{=}\left({}^{(6.2)}\mathbf{E}^{\alpha}_{\mathbf{0n}}\right)_{(M,\mu)(N,\nu)}}\Bigg\}
\end{aligned}
\tag{3.53}
$$

with

$$\underbrace{({}^{(6.1)}\mathbf{E}^{\alpha}_{\mathbf{0n}})_{(M,\mu)(N,\nu)}}_{\mathbf{n}N\neq\mathbf{0}M} = \sum_{\mathbf{l}=\mathbf{N}^-}^{\mathbf{N}^+}\sum_{L=1}^{N_n}\sum_{\tau=1}^{n_o(N)}\sum_{\lambda=1}^{n_o(L)}(\mathbf{P}^{\alpha}_{\mathbf{0l}})_{(N,\tau)(L,\lambda)}\sum_{\mu'=1}^{n_o(M)}\left(\begin{array}{cc}\mathbf{0} & \mathbf{l}\\ M & L\\ \mu' & \lambda\end{array}\right)\left(\begin{array}{cc|cc}\mathbf{0} & \mathbf{n} & \mathbf{n} & \mathbf{0}\\ M & N & N & M\\ \mu & \tau & \nu & \mu'\end{array}\right)$$
$$-\sum_{\tau=1}^{n_o(N)}\sum_{\lambda=1}^{n_o(N)}(\mathbf{P}^{\alpha}_{\mathbf{00}})_{(N,\tau)(N,\lambda)}\sum_{\mu'=1}^{n_o(M)}\left(\begin{array}{cc}\mathbf{0} & \mathbf{n}\\ M & N\\ \mu' & \lambda\end{array}\right)\left(\begin{array}{cc|cc}\mathbf{0} & \mathbf{n} & \mathbf{n} & \mathbf{0}\\ M & N & N & M\\ \mu & \tau & \nu & \mu'\end{array}\right) \tag{3.54}$$
$$-\sum_{\tau=1}^{n_o(N)}\sum_{\lambda=1}^{n_o(M)}(\mathbf{P}^{\alpha}_{\mathbf{0n}})_{(M,\lambda)(N,\tau)}\left(\begin{array}{cc|cc}\mathbf{0} & \mathbf{n} & \mathbf{n} & \mathbf{0}\\ M & N & N & M\\ \mu & \tau & \nu & \lambda\end{array}\right)$$

and

$$\underbrace{({}^{(6.2)}\mathbf{E}^{\alpha}_{\mathbf{0n}})_{(M,\mu)(N,\nu)}}_{\mathbf{n}N\neq\mathbf{0}M} = \underbrace{\sum_{\mathbf{l}=\mathbf{N}^-}^{\mathbf{N}^+}\sum_{L=1}^{N_n}}_{\mathbf{l}L\neq\mathbf{0}N}\sum_{\tau=1}^{n_o(N)}\sum_{\lambda=1}^{n_o(L)}(\mathbf{P}^{\alpha}_{\mathbf{0l}})_{(N,\tau)(L,\lambda)}\sum_{\lambda'=1}^{n_o(L)}\left(\begin{array}{cc}\mathbf{0} & \mathbf{l}\\ M & L\\ \mu & \lambda'\end{array}\right)\left(\begin{array}{cc|cc}\mathbf{l} & \mathbf{0} & \mathbf{0} & \mathbf{l}\\ L & N & N & L\\ \lambda' & \tau & \nu & \lambda\end{array}\right)$$
$$-\sum_{\tau=1}^{n_o(N)}\sum_{\lambda=1}^{n_o(N)}(\mathbf{P}^{\alpha}_{\mathbf{00}})_{(N,\tau)(N,\lambda)}\sum_{\lambda'=1}^{n_o(N)}\left\{\left(\begin{array}{cc}\mathbf{0} & \mathbf{n}\\ M & N\\ \mu & \lambda'\end{array}\right)-\left(\begin{array}{cc}\mathbf{0} & \mathbf{0}\\ M & N\\ \mu & \lambda'\end{array}\right)\right\}\left(\begin{array}{cc|cc}\mathbf{0} & \mathbf{0} & \mathbf{0} & \mathbf{0}\\ N & N & N & N\\ \lambda' & \tau & \nu & \lambda\end{array}\right) \tag{3.55}$$
$$-\sum_{\tau=1}^{n_o(N)}\sum_{\lambda=1}^{n_o(M)}(\mathbf{P}^{\alpha}_{\mathbf{0n}})_{(M,\lambda)(N,\tau)}\left(\begin{array}{cc|cc}\mathbf{0} & \mathbf{n} & \mathbf{n} & \mathbf{0}\\ M & N & N & M\\ \mu & \tau & \nu & \lambda\end{array}\right).$$

4. Real-space Lattice Summations Including Long-range Interactions

4.1. Long-range Correction of the Fock Matrix

We now return to the problem mentioned above in connection with ([6] 2.19) and ([6] 2.20) concerning real-space lattice sums. With respect to ([6] 2.19), the choice of unit-cell numbers N_a, N_b, and N_c, which governs the expense of real-space lattice summations in general, can be kept small, since the matrices $(\mathbf{S}_{\mathbf{0n}})$ decay rather quickly with increasing intercellular distances $|\mathbf{R_0}-\mathbf{R_n}|$. The Fock-matrix representation of ([6] 2.20), on the other hand, contains long-ranging two-center integrals of Coulomb type. For large internuclear distances these Coulomb-type attraction and repulsion integrals can be identified approximately with classical Coulomb interaction energies:

$$\left(\begin{array}{ccc}\mathbf{0} & \mathbf{t} & \mathbf{0}\\ M & T & M\\ \mu & & \mu\end{array}\right)_{\infty} := -Z_T|\mathbf{R}_M-[\mathbf{R}_T-\mathbf{R_t}]|^{-1} \text{ for all } \mu, \tag{4.1}$$

$$\left(\begin{array}{cc|cc}\mathbf{0} & \mathbf{0} & \mathbf{t} & \mathbf{t}\\ M & M & T & T\\ \mu & \mu & \tau & \tau\end{array}\right)_{\infty} := |\mathbf{R}_M-[\mathbf{R}_T-\mathbf{R_t}]|^{-1} \text{ for all } \mu,\tau. \tag{4.2}$$

Assuming that all integrals except those of (4.1) and (4.2) have decayed sufficiently, if

$$(\mathbf{S}_{\mathbf{0}\{N_a^+,0,0\}}) \approx (\mathbf{S}_{\mathbf{0}\{0,N_b^+,0\}}) \approx (\mathbf{S}_{\mathbf{0}\{0,0,N_c^+\}}) \approx \mathbf{0}, \tag{4.3}$$

we additionally have to specify the following long-range corrections. Using the atomic charge definition for a locally orthonormalized basis set

$$q_M^{\infty} \stackrel{\text{def}}{=} Z_M - \sum_{\mu=1}^{n_o(M)}(\mathbf{P}^{\oplus}_{\mathbf{00}})_{(M,\mu)(M,\mu)}, \tag{4.4}$$

we distinguish two off-diagonal cases and the diagonal one. For the diatomic off-diagonal case we write

$$\underbrace{F'^{\alpha}_{(M,\mu)(N,\nu)}(\mathbf{k})}_{N\neq M} :=$$
$$\sum_{\mathbf{n}=\mathbf{N}^-}^{\mathbf{N}^+}\exp(i[k^a n_a|\mathbf{a}|+k^b n_b|\mathbf{b}|+k^c n_c|\mathbf{c}|])(\mathbf{F}^{\alpha}_{\mathbf{0n}})_{(M,\mu)(N,\nu)}$$
$$+F'^{\alpha\infty}_{(M,\mu)(N,\nu)}(\mathbf{k}) - F'^{\alpha\circ}_{(M,\mu)(N,\nu)}(\mathbf{k}). \tag{4.5}$$

From the (2.1) … (2.8) we pick out all long-range terms and collect them in $F'^{\alpha\infty}_{(M,\mu)(N,\nu)}(\mathbf{k})$:

$$\underbrace{F'^{\alpha\infty}_{(M,\mu)(N,\nu)}(\mathbf{k})}_{N\neq M} \stackrel{\text{def}}{=} -\sum_{\mathbf{n}=-\boldsymbol{\infty}}^{+\boldsymbol{\infty}} \exp(i[k^a n_a|\mathbf{a}|+k^b n_b|\mathbf{b}|+k^c n_c|\mathbf{c}|])(\mathbf{P}^{\alpha}_{\mathbf{0n}})_{(M,\mu)(N,\nu)} \underbrace{\left(\begin{array}{cc|cc} \mathbf{0} & \mathbf{0} & \mathbf{n} & \mathbf{n} \\ M & M & N & N \\ \mu & \mu & \nu & \nu \end{array}\right)_{\infty}}_{:=|\mathbf{R}_M-[\mathbf{R}_N-\mathbf{R}_\mathbf{n}]|^{-1}}. \tag{4.6}$$

Using ([6] 2.25), we rewrite

$$(\mathbf{P}^{\alpha}_{\mathbf{0n}})_{(M,\mu)(N,\nu)} := \frac{V_d}{(2\pi)^d}\int\!\!\int\!\!\int \exp\Big\{-i\left[k'^a n_a|\mathbf{a}|+k'^b n_b|\mathbf{b}|+k'^c n_c|\mathbf{c}|\right]\Big\} P'^{\alpha}_{(M,\mu)(N,\nu)}(\mathbf{k}')\mathrm{d}k'^a\mathrm{d}k'^b\mathrm{d}k'^c. \tag{4.7}$$

Inserting (4.7) into (4.6) we get

$$\begin{aligned}\underbrace{F'^{\alpha\infty}_{(M,\mu)(N,\nu)}(\mathbf{k})}_{N\neq M} &:= -\frac{V_d}{(2\pi)^d}\int\!\!\int\!\!\int \sum_{\mathbf{n}=-\boldsymbol{\infty}}^{+\boldsymbol{\infty}} \exp\Big\{i[(k^a-k'^a)n_a|\mathbf{a}|+(k^b-k'^b)n_b|\mathbf{b}|+(k^c-k'^c)n_c|\mathbf{c}|]\Big\} \\ &\qquad\times P'^{\alpha}_{(M,\mu)(N,\nu)}(\mathbf{k}')\mathrm{d}k'^a\mathrm{d}k'^b\mathrm{d}k'^c|\mathbf{R}_M-[\mathbf{R}_N-\mathbf{R}_\mathbf{n}]|^{-1} \\ &= -P'^{\alpha}_{(M,\mu)(N,\nu)}(\mathbf{0})\sum_{\mathbf{n}=-\boldsymbol{\infty}}^{+\boldsymbol{\infty}} \exp\Big(i[k^a n_a|\mathbf{a}|+k^b n_b|\mathbf{b}|+k^c n_c|\mathbf{c}|]\Big)|\mathbf{R}_M-[\mathbf{R}_N-\mathbf{R}_\mathbf{n}]|^{-1}.\end{aligned} \tag{4.8}$$

$F'^{\alpha\circ}_{(M,\mu)(N,\nu)}(\mathbf{k})$ represents the sum of those classical contributions which had already been included non-classically:

$$\underbrace{F'^{\alpha\circ}_{(M,\mu)(N,\nu)}(\mathbf{k})}_{N\neq M} \stackrel{\text{def}}{=} -\sum_{\mathbf{n}=\mathbf{N}^-}^{\mathbf{N}^+} \exp\Big(i[k^a n_a|\mathbf{a}|+k^b n_b|\mathbf{b}|+k^c n_c|\mathbf{c}|]\Big)\times(\mathbf{P}^{\alpha}_{\mathbf{0n}})_{(M,\mu)(N,\nu)}|\mathbf{R}_M-[\mathbf{R}_N-\mathbf{R}_\mathbf{n}]|^{-1} \tag{4.9}$$

or according to (4.8)

$$\underbrace{F'^{\alpha\circ}_{(M,\mu)(N,\nu)}(\mathbf{k})}_{N\neq M} = -P'^{\alpha}_{(M,\mu)(N,\nu)}(\mathbf{0})\sum_{\mathbf{n}=\mathbf{N}^-}^{\mathbf{N}^+} \exp\Big(i[k^a n_a|\mathbf{a}|+k^b n_b|\mathbf{b}|+k^c n_c|\mathbf{c}|]\Big)\times|\mathbf{R}_M-[\mathbf{R}_N-\mathbf{R}_\mathbf{n}]|^{-1}. \tag{4.10}$$

For the one-center off-diagonal case we simply get

$$\underbrace{F'^{\alpha}_{(M,\mu)(M,\nu)}(\mathbf{k})}_{\nu\neq\mu} := \sum_{\mathbf{n}=\mathbf{N}^-}^{\mathbf{N}^+} \exp\Big(i[k^a n_a|\mathbf{a}|+k^b n_b|\mathbf{b}|+k^c n_c|\mathbf{c}|]\Big)(\mathbf{F}^{\alpha}_{\mathbf{0n}})_{(M,\mu)(M,\nu)}. \tag{4.11}$$

For the diagonal elements we define

$$F'^{\alpha}_{(M,\mu)(M,\mu)}(\mathbf{k}) := \sum_{\mathbf{n}=\mathbf{N}^-}^{\mathbf{N}^+} \exp\Big(i[k^a n_a|\mathbf{a}|+k^b n_b|\mathbf{b}|+k^c n_c|\mathbf{c}|]\Big)\times(\mathbf{F}^{\alpha}_{\mathbf{0n}})_{(M,\mu)(M,\mu)}+F^{\infty}_{(M,\mu)(M,\mu)}-F^{\circ}_{(M,\mu)(M,\mu)}. \tag{4.12}$$

Again, from the (2.1) … (2.8) we pick out all long-range terms and collect them in $F^{\ \infty}_{(M,\mu)(M,\mu)}$:

$$\begin{aligned} F^{\infty}_{(M,\mu)(M,\mu)} &\stackrel{\text{def}}{=} \underbrace{\sum_{\mathbf{t}=-\boldsymbol{\infty}}^{+\boldsymbol{\infty}}\sum_{T=1}^{N_n}}_{\mathbf{t}T\neq\mathbf{0}M}\Bigg\{\underbrace{\begin{pmatrix} \mathbf{0} & \mathbf{t} & \mathbf{0} \\ M & T & M \\ \mu & & \mu \end{pmatrix}_{\infty}}_{:=-Z_T|\mathbf{R}_M-[\mathbf{R}_T-\mathbf{R}_\mathbf{t}]|^{-1}} + \underbrace{\sum_{\tau=1}^{n_o(T)}(\mathbf{P}^{\oplus}_{\mathbf{00}})_{(T,\tau)(T,\tau)}}_{:=Z_T-q^{\infty}_T}\underbrace{\left(\begin{array}{cc|cc} \mathbf{0} & \mathbf{0} & \mathbf{t} & \mathbf{t} \\ M & M & T & T \\ \mu & \mu & \tau & \tau \end{array}\right)_{\infty}}_{:=|\mathbf{R}_M-[\mathbf{R}_T-\mathbf{R}_\mathbf{t}]|^{-1}}\Bigg\} \\ &= \underbrace{\sum_{\mathbf{t}=-\boldsymbol{\infty}}^{+\boldsymbol{\infty}}\sum_{T=1}^{N_n}}_{\mathbf{t}T\neq\mathbf{0}M}(-q^{\infty}_T)|\mathbf{R}_M-[\mathbf{R}_T-\mathbf{R}_\mathbf{t}]|^{-1} \quad \text{for all } \mu. \end{aligned} \tag{4.13}$$

Correspondingly, we write

$$F^{\circ}_{(M,\mu)(M,\mu)} \overset{\text{def}}{=} \underbrace{\sum_{\mathbf{t}=\mathbf{N}^-}^{\mathbf{N}^+} \sum_{T=1}^{N_n}}_{\mathbf{t}T\neq\mathbf{0}M} (-q_T)|\mathbf{R}_M - [\mathbf{R}_T - \mathbf{R}_\mathbf{t}]|^{-1} \quad \text{for all } \mu. \tag{4.14}$$

Following Ramírez and Böhm [14], (4.13) can be interpreted as follows : For large internuclear distances each non-zero Fock-matrix element $F^{\infty}_{(M,\mu)(M,\mu)}$ belonging to an atom M represents the Madelung potential of an electronic point charge at the center M of the reference cell. Such Madelung potentials can be evaluated using Ewald-summation techniques [16].

Off-diagonal contributions, however, which stem from long-range exchange interactions, have no classical counterpart in our description. Since the infinite sums of (4.6) are only conditionally convergent, their evaluation causes severe problems in solid-state Hartree-Fock theories [17]. Numerous techniques and recipes have been proposed to overcome such difficulties (cf. [18] and references therein).

4.2. Long-range Correction of the Total Energy per Unit Cell

Finally, we discuss real-space lattice summations in the computation of the total energy per unit cell:

$$\begin{aligned} \mathcal{E}_t = \frac{1}{2} \sum_{M=1}^{N_n} \sum_{\mu=1}^{n_o(M)} \sum_{\mathbf{n}=\mathbf{N}^-}^{\mathbf{N}^+} \sum_{N=1}^{N_n} \sum_{\nu=1}^{n_o(N)} \Big\{ (\mathbf{P}^{\oplus}_{\mathbf{0n}})_{(M,\mu)(N,\nu)} \Big[(\mathbf{K}_{\mathbf{0n}})_{(M,\mu)(N,\nu)} + (\mathbf{F}^{A}_{\mathbf{0n}})_{(M,\mu)(N,\nu)} \Big] \\ + (\mathbf{P}^{\alpha}_{\mathbf{0n}})_{(M,\mu)(N,\nu)} (\mathbf{F}^{\alpha}_{\mathbf{0n}})_{(M,\mu)(N,\nu)} + (\mathbf{P}^{\beta}_{\mathbf{0n}})_{(M,\mu)(N,\nu)} (\mathbf{F}^{\beta}_{\mathbf{0n}})_{(M,\mu)(N,\nu)} \Big\} \\ + \underbrace{\frac{1}{2} \sum_{M=1}^{N_n} Z_M \underbrace{\sum_{\mathbf{n}=\mathbf{N}^-}^{\mathbf{N}^+} \sum_{N=1}^{N_n}}_{\mathbf{n}N\neq\mathbf{0}M} Z_N |\mathbf{R}_M - [\mathbf{R}_N - \mathbf{R}_\mathbf{n}]|^{-1}}_{\overset{\text{def}}{=}\mathcal{E}_n} + \mathcal{E}_{t\infty} - \mathcal{E}_{t\circ}. \end{aligned} \tag{4.15}$$

According to ([6] 2.24) we can also write

$$\begin{aligned} \mathcal{E}_t = \frac{1}{2} \sum_{M=1}^{N_n} \sum_{\mu=1}^{n_o(M)} \sum_{\mathbf{n}=\mathbf{N}^-}^{\mathbf{N}^+} \sum_{N=1}^{N_n} \sum_{\nu=1}^{n_o(N)} \Big\{ (\mathbf{P}^{\oplus}_{\mathbf{0n}})_{(M,\mu)(N,\nu)} \Big[2(\mathbf{K}_{\mathbf{0n}})_{(M,\mu)(N,\nu)} + 2(\mathbf{F}^{A}_{\mathbf{0n}})_{(M,\mu)(N,\nu)} + (\mathbf{F}^{C}_{\mathbf{0n}})_{(M,\mu)(N,\nu)} \Big] \\ - (\mathbf{P}^{\alpha}_{\mathbf{0n}})_{(M,\mu)(N,\nu)} (\mathbf{F}^{\alpha E}_{\mathbf{0n}})_{(M,\mu)(N,\nu)} - (\mathbf{P}^{\beta}_{\mathbf{0n}})_{(M,\mu)(N,\nu)} (\mathbf{F}^{\beta E}_{\mathbf{0n}})_{(M,\mu)(N,\nu)} \Big\} \\ + \mathcal{E}_n + \mathcal{E}_{t\infty} - \mathcal{E}_{t\circ}. \end{aligned} \tag{4.16}$$

As before, we pick out all long-range terms from $\mathcal{E}_n$ and (2.1) ... (2.8), and collect them in $\mathcal{E}_{t\infty}$:

$$\mathcal{E}_{t\infty} \overset{\text{def}}{=} \frac{1}{2} \sum_{M=1}^{N_n} Z_M \underbrace{\sum_{\mathbf{n}=-\infty}^{+\infty} \sum_{N=1}^{N_n}}_{\mathbf{n}N\neq\mathbf{0}M} Z_N |\mathbf{R}_M - [\mathbf{R}_N - \mathbf{R}_\mathbf{n}]|^{-1} + \sum_{M=1}^{N_n} \underbrace{\sum_{\mu=1}^{n_o(M)} (\mathbf{P}^{\oplus}_{\mathbf{00}})_{(M,\mu)(M,\mu)}}_{:=(Z_M - q_M)} \underbrace{\sum_{\mathbf{n}=-\infty}^{+\infty} \sum_{N=1}^{N_n}}_{\mathbf{n}N\neq\mathbf{0}M} \underbrace{\begin{pmatrix} \mathbf{0} & \mathbf{n} & \mathbf{0} \\ M & N & M \\ \mu & & \mu \end{pmatrix}_{\infty}}_{:=-Z_N|\mathbf{R}_M - [\mathbf{R}_N - \mathbf{R}_\mathbf{n}]|^{-1}}$$

$$
\begin{aligned}
&+\frac{1}{2}\sum_{M=1}^{N_n}\underbrace{\sum_{\mu=1}^{n_o(M)}(\mathbf{P}^{\oplus}_{\mathbf{00}})_{(M,\mu)(M,\mu)}}_{:=(Z_M-q_M)}\underbrace{\sum_{\mathbf{n}=-\infty}^{+\infty}\sum_{N=1}^{N_n}}_{\mathbf{n}N\neq\mathbf{0}M}\underbrace{\sum_{\nu=1}^{n_o(N)}(\mathbf{P}^{\oplus}_{\mathbf{00}})_{(N,\nu)(N,\nu)}}_{:=(Z_N-q_N)}\underbrace{\left(\begin{array}{cc|cc}\mathbf{0}&\mathbf{0}&\mathbf{n}&\mathbf{n}\\ M&M&N&N\\ \mu&\mu&\nu&\nu\end{array}\right)_{\infty}}_{:=|\mathbf{R}_M-[\mathbf{R}_N-\mathbf{R}_\mathbf{n}]|^{-1}}\\
&-\frac{1}{2}\sum_{M=1}^{N_n}\sum_{\mu=1}^{n_o(M)}\underbrace{\sum_{\mathbf{n}=-\infty}^{+\infty}\sum_{N=1}^{N_n}}_{\mathbf{n}N\neq\mathbf{0}M}\sum_{\nu=1}^{n_o(N)}(\mathbf{P}^{\alpha}_{\mathbf{0n}})^2_{(M,\mu)(N,\nu)}\underbrace{\left(\begin{array}{cc|cc}\mathbf{0}&\mathbf{0}&\mathbf{n}&\mathbf{n}\\ M&M&N&N\\ \mu&\mu&\nu&\nu\end{array}\right)_{\infty}}_{:=|\mathbf{R}_M-[\mathbf{R}_N-\mathbf{R}_\mathbf{n}]|^{-1}}\\
&-\frac{1}{2}\sum_{M=1}^{N_n}\sum_{\mu=1}^{n_o(M)}\underbrace{\sum_{\mathbf{n}=-\infty}^{+\infty}\sum_{N=1}^{N_n}}_{\mathbf{n}N\neq\mathbf{0}M}\sum_{\nu=1}^{n_o(N)}(\mathbf{P}^{\beta}_{\mathbf{0n}})^2_{(M,\mu)(N,\nu)}\underbrace{\left(\begin{array}{cc|cc}\mathbf{0}&\mathbf{0}&\mathbf{n}&\mathbf{n}\\ M&M&N&N\\ \mu&\mu&\nu&\nu\end{array}\right)_{\infty}}_{:=|\mathbf{R}_M-[\mathbf{R}_N-\mathbf{R}_\mathbf{n}]|^{-1}}\\
&=\underbrace{\frac{1}{2}\sum_{M=1}^{N_n}q_M^{\infty}\underbrace{\sum_{\mathbf{n}=-\infty}^{+\infty}\sum_{N=1}^{N_n}}_{\mathbf{n}N\neq\mathbf{0}M}q_N^{\infty}|\mathbf{R}_M-[\mathbf{R}_N-\mathbf{R}_\mathbf{n}]|^{-1}}_{\stackrel{\text{def}}{=}\mathcal{E}^{\mathcal{M}}_{t\infty}}+\mathcal{E}^{E}_{t\infty},
\end{aligned}
\tag{4.17}
$$

where

$$
\mathcal{E}^{E}_{t\infty}\stackrel{\text{def}}{=}-\frac{1}{2}\sum_{M=1}^{N_n}\underbrace{\sum_{\mathbf{n}=-\infty}^{+\infty}\sum_{N=1}^{N_n}}_{\mathbf{n}N\neq\mathbf{0}M}|\mathbf{R}_M-[\mathbf{R}_N-\mathbf{R}_\mathbf{n}]|^{-1}\sum_{\mu=1}^{n_o(M)}\sum_{\nu=1}^{n_o(N)}\left[(\mathbf{P}^{\alpha}_{\mathbf{0n}})^2_{(M,\mu)(N,\nu)}+(\mathbf{P}^{\beta}_{\mathbf{0n}})^2_{(M,\mu)(N,\nu)}\right]
\tag{4.18}
$$

and

$$
\mathcal{E}_{t\circ}\stackrel{\text{def}}{=}\underbrace{\frac{1}{2}\sum_{M=1}^{N_n}q_M\underbrace{\sum_{\mathbf{n}=\mathbf{N}^-}^{\mathbf{N}^+}\sum_{N=1}^{N_n}}_{\mathbf{n}N\neq\mathbf{0}M}q_N|\mathbf{R}_M-[\mathbf{R}_N-\mathbf{R}_\mathbf{n}]|^{-1}}_{\stackrel{\text{def}}{=}\mathcal{E}^{\mathcal{M}}_{t\circ}}+\mathcal{E}^{E}_{t\circ}
\tag{4.19}
$$

with

$$
\mathcal{E}^{E}_{t\circ}\stackrel{\text{def}}{=}-\frac{1}{2}\sum_{M=1}^{N_n}\underbrace{\sum_{\mathbf{n}=\mathbf{N}^-}^{\mathbf{N}^+}\sum_{N=1}^{N_n}}_{\mathbf{n}N\neq\mathbf{0}M}|\mathbf{R}_M-[\mathbf{R}_N-\mathbf{R}_\mathbf{n}]|^{-1}\sum_{\mu=1}^{n_o(M)}\sum_{\nu=1}^{n_o(N)}\left[(\mathbf{P}^{\alpha}_{\mathbf{0n}})^2_{(M,\mu)(N,\nu)}+(\mathbf{P}^{\beta}_{\mathbf{0n}})^2_{(M,\mu)(N,\nu)}\right].
\tag{4.20}
$$

Again, $\mathcal{E}^{\mathcal{M}}_{t\infty}$ is the Madelung energy of a crystalline system with point charges q_M at $\mathbf{R}_M$ and q_N at $(\mathbf{R}_N-\mathbf{R}_n)$. It can also be evaluated by means of Ewald-summation techniques [16]. How the infinite sum of (4.18) can be treated, which arises from long-range off-diagonal exchange contributions, is again a hard problem of any *ab-initio* solid-state theory of Hartree-Fock type (cf. [18] and references therein) [19].

5. Concluding Remarks

The present contribution shows that the insights of the preceding paper obtained for the molecular case [1] can be extended to a related quantum chemical solid-state formalism.

For practical purposes, we here are particularly interested in the R.R&C branch which requires a computational procedure for the accurate evaluation of all two-center integrals.

Due to their dependence on a single geometric parameter, all types of two-center integrals can be calculated in advance for about one hundred fixed interatomic distances at the desired level of sophistication [20] and stored once and for all. A cubic spline algorithm [21] may be taken to interpolate the actual integral value from each precomputed list. Such techniques, particularly appropriate for minimal basis sets, have been incorporated in a non-empirical crystal orbital procedure based on Rüdenberg's ideas.

Acknowledgement (W.K.)

About twentyfive years ago, I first became acquainted with the theory of (one-dimensional) crystals through Professor Seelig's quantum chemical lectures. Later on he invited me to join his working group to perform band-structure calculations at the "Extended-Hückel" level which seemed to be appropriate for our purposes and computer facilities at that time. While conserving its conceptual simplicity, my scientific efforts of the following years intended to improve this simple semi-empirical picture. The distilled result, which is outlined in the present contribution, appears as a non-empirical variant of Professor Seelig's early concept [22].

I would like to express my gratitude for the many years of cooperation in our small department for Theoretische Chemie, together with sincere congratulations that include best wishes for our Professor's and his family's future.

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[17] In the U&C variant described in a previous paper [6], however, the long-range contribution of (4.6) disappears completely:

$$\underbrace{F'^{\alpha\infty[\mathrm{R.U\&C}]}_{(M,\mu)(N,\nu)}(\mathbf{k})}_{N\neq M} - \underbrace{F'^{\alpha\circ[\mathrm{R.U\&C}]}_{(M,\mu)(N,\nu)}(\mathbf{k})}_{N\neq M} := 0.$$

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[19] In the already mentioned U&C variant [6], however, the long-range contribution of (4.16) again disappears:

$$\mathcal{E}^{E[\mathrm{R.U\&C}]}_{t\infty} - \mathcal{E}^{E[\mathrm{R.U\&C}]}_{t\circ} := 0.$$

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