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Published on: 01 Apr 1996 - Journal of Chemical Physics (American Institute of Physics)

Topics: Tensor operator and Shift operator

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Citation: The Journal of Chemical Physics **104**, 5112 (1996); doi: 10.1063/1.471139
View online: http://dx.doi.org/10.1063/1.471139
View Table of Contents: http://scitation.aip.org/content/aip/journal/jcp/104/13?ver=pdfcov
Published by the AIP Publishing

Matrix elements of $U(2n)$ generators in a multishell spin–orbit basis. I. General formalism

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(Received 10 May 1995; accepted 20 December 1995)

This is the first in a series of papers which derives the matrix elements of the spin-dependent $U(2n)$ generators in a multishell spin–orbit basis, i.e., a spin adapted composite Gelfand–Paldus basis. The advantages of such a multishell formalism are well known and well documented. The approach taken exploits the properties of the $U(n)$ adjoint tensor operator denoted by $\Delta^i_j(1 \leqslant i,j \leqslant n)$ as defined by Gould and Paldus [J. Chem. Phys. **92**, 7394 (1990)]. Δ is a polynomial of degree two in the $U(n)$ matrix $E=[E^i_j]$. The unique properties of this operator allow the construction of adjoint coupling coefficients for the zero-shift components of the $U(2n)$ generators. The Racah factorization lemma may then be applied to obtain the matrix elements of all the $U(2n)$ generators. In this paper we investigate the underlying formalism of the approach and discuss its advantages and its relationship to the shift operator method of Gould and Battle [J. Chem. Phys. **99**, 5961 (1993)]. The formalism is then applied, in the second paper of the series, to calculate the matrix elements of the del operator in a two-shell spin–orbit basis. This immediately yields the zero-shift adjoint coupling coefficients in such a basis. The del-operator matrix elements are required for the calculation of spin densities in a two-shell basis. In the third paper of the series we derive the remaining nonzero shift adjoint coupling coefficients all of which are required for the multishell case. We then use these coupling coefficients to obtain formulas for the matrix elements of the $U(2n)$ generators in a two-shell spin–orbit basis. This result is then generalized, in the fourth paper, to the case of the multishell spin–orbit basis. Finally, we demonstrate that in the Gefand–Tsetlin limit the formula obtained is equivalent to that of Gould and Battle for a single-shell system. [S0021-9606(96)00912-3]

I. INTRODUCTION

We present in this series of papers a derivation of the matrix elements (MEs) of the generators of the spin–orbital unitary group $U(2n)$ in a multishell spin–orbit basis. Our works extends that of Gould and Paldus[1] and Gould and Battle[2] who obtained $U(2n)$ generator MEs for the single-shell case. It is based entirely on the $U(n)$ representation theory developed for the many electron problem by Paldus[3] and extended by Gould *et al.*[4–11]

The unitary group approach (UGA) to configuration interaction (CI) is motivated by the fact that the spin-independent molecular electronic Hamiltonian is expressible in terms of the $U(n)$ generators, viz.

$$H_0=\sum_{i,j=1}^{n} \langle i|\hat{z}|j\rangle E^i_j+\frac{1}{2}\sum_{i,j,k,l=1}^{n}(ij|kl)(E^i_jE^k_l-\delta^k_jE^i_l). \tag{1}$$

Here, the coefficients $\langle i|\hat{z}|j\rangle$ and $(ij|kl)$ are one-electron and two-electron integrals, respectively,[12] and E^i_j denotes the generator of the orbital unitary group $U(n)$.

This fact gives rise to the need to calculate the MEs of the $U(n)$ generators. The appropriate basis is the Gelfand–Paldus (GP) basis which is the Gelfand–Tsetlin (GT) basis for the many-electron problem.[3] This is a basis defined by symmetry adaptation to the canonical chain of subgroups

$$U(n)\supset U(n-1)\supset\cdots\supset U(2)\supset U(1). \tag{2}$$

The labeling information thus obtained together with the Pauli exclusion principle, provides all the information necessary to describe a spin-independent molecular state.

When spin-dependent effects are to be included in the Hamiltonian extra terms involving the $U(2n)$ generators must be added. Now we have, for the case of one-body spin-dependent interactions,

$$H=H_0+H_S, \tag{3}$$

where

$$H_S=\sum_{i,j=1}^{n}\sum_{\mu,\nu=0}^{1}\langle i\mu|H_S|j\nu\rangle E^{i\mu}_{j\nu}. \tag{4}$$

As for the above, the coefficient $\langle i\mu|H_S|j\nu\rangle$ is a one-electron integral and $E^{i\mu}_{j\nu}$ is the $U(2n)$ generator.

Now, for the spin-dependent problem, a more appropriate basis to work with is one which is symmetry adapted to the subgroup chain

$$U(2n)\supset U(n)\times U(2). \tag{5}$$

This is referred to as the spin–orbit (SO) basis.[1] The right-hand side is the outer direct product of the orbital group $U(n)$ and the spin group $U(2)$. We now need to calculate the MEs of the $U(2n)$ generators in this spin–orbit basis. The effect of the $U(2n)$ generators may be analyzed according as to how they affect the total spin, S, of the state upon which they act.

Now, the $U(2n)$ generators transform under commutation with the $U(n)\times U(2)$ generators as the representation $\mathrm{Adj}[U(n)]\otimes\mathrm{Adj}[U(2)]$. Here, $\mathrm{Adj}[G]$ denotes the adjoint representation of the group G. This means that the $U(2n)$ generators are adjoint tensor operators (ATOs) of both $U(n)$ and $U(2)$, and in particular it is true of any ATO of $U(2)$ that it may be resolved into spin shift components.[1] Thus we may write

$$E^{i\mu}_{j\nu}=E(+)^{i\mu}_{j\nu}+E(0)^{i\mu}_{j\nu}+E(-)^{i\mu}_{j\nu}, \tag{6}$$

where $E(\pm)^{i\mu}_{j\nu}$, respectively, increases/decreases the spin S by one unit and $E(0)^{i\mu}_{j\nu}$ leaves the spin unaltered.

For the nonzero spin shift components, $E(\pm)^{i\mu}_{j\nu}$, the MEs may be determined in a straightforward manner by applying the $U(n)\times U(2)$ Wigner–Eckart (WE) theorem.[1] However, for the zero-shift components, $E(0)^{i\mu}_{j\nu}$, this cannot be done.

From the work of Louck and Biedenharn[13] on the general theory of ATOs, it is known that there exist several independent zero-shift adjoint tensors. These act as a basis set for the general ATO which can be expressed as a linear combination of them. Due to the multiplicity involved, in the general case (that is, for an arbitrary representation) there is no unique set of coupling coefficients. Thus the WE theorem cannot be applied in its usual form.

As we will demonstrate below, for the many electron problem it is in fact possible to define a set of zero-shift adjoint coupling coefficients. This is accomplished via the $U(n)$ operator Δ which is a polynomial of degree two in the $U(n)$ matrix $E=[E^i_j]$ $(1\leq i,j\leq n)$. The del operator was introduced by Gould and Paldus in Ref. 1. They obtained ME formulas in terms of it for both the zero and nonzero shift components of the $U(2n)$ generators. The del operator is defined by

$$\Delta^i_j=E(E+N/2-n-2)^i_j. \tag{7}$$

The MEs of this operator are important not just for spin-dependent CI but, as was shown by Gould, Paldus, and Chandler, they are also required for obtaining molecular electronic spin densities.[14]

The del operator possesses unique properties which, for the many electron problems, make possible the construction of appropriate zero-shift adjoint coupling coefficients. We discuss the del operator in Sec. IV. These coupling coefficients are essentially the MEs of the normalized del operators. That is, we *define* (for the single shell case)

$$\left\langle \begin{matrix} p \\ P' \end{matrix} \middle| e_i\otimes\bar{e}_j; \begin{matrix} p \\ P \end{matrix} \right\rangle = \frac{-1}{2S(S+1)} \left\langle \begin{matrix} p \\ P' \end{matrix} \middle| \Delta^i_j \middle| \begin{matrix} p \\ P \end{matrix} \right\rangle. \tag{8}$$

Here and throughout it is understood that the lhs vanishes if $S=0$ (which is consistent with the fact that $\Delta^i_j=0$ in such a case[14]). The nonzero shift adjoint coupling coefficients (ACCs) are, of course, uniquely defined.

By means of the above definition we may now adopt a unified approach to the evaluation of the $U(2n)$ generator matrix elements. By the application of the Racah factorization lemma, we express the MEs of both the zero and nonzero shift components in terms of adjoint coupling coefficients and reduced Wigner coefficients (RWCs).

This adjoint coupling coefficient approach is considerably simpler than the shift component method (SCM) of Gould and Battle in Ref. 2. We demonstrate later in this paper that it yields the same formulae for the del-operator matrix elements. The aim of this first paper in the series is to develop the formalism of the adjoint coupling coefficient approach and to demonstrate its equivalence to the shift component method for the one-shell case. In the remaining papers of the series we show that the method can be extended to the multishell case and we obtain formulas for the $U(2n)$ generator MEs first in a two-shell spin–orbit basis and then in the general multishell spin–orbit basis.

The need for a multishell formalism arises from the fact that the electrons of a molecular system can sometimes be separated into well defined groups or shells.[8] This is due to the localization of the molecular orbitals around constituent atoms, the effect of symmetry constraints, or energy effects. The quantum chemistry model needs to be extended to systems where, in addition, spin effects are also important. In the spin-dependent shell model, where we have shell j with N_j electrons possessing a definite spin S_j, the spins of the separate shells couple to a total spin S and the set of shells together form a configuration

$$({}^{S_1}N_1,{}^{S_2}N_2,\ldots,{}^{S_r}N_r), \quad \left(\sum_{i=1}^{r} N_i=N\right). \tag{9}$$

For such a system we choose a basis which is symmetry adapted to the subgroup chain

$$\begin{aligned} U(2n) &\supset U(2n_1)\times U(2n_2)\times\cdots\times U(2n_r) \\ &\supset\cdots \\ &\supset U(n_1)\times U(n_2)\times\cdots\times U(n_r)\times U(2) \end{aligned} \tag{10}$$

and refer to this as the multishell spin–orbit basis. Our aim in this series is to obtain the $U(2n)$ generator MEs in this basis.

This, the first paper of the series is set up as follows. The basic notation and terminology, together with a review of frequently used standard theory (such as the Wigner–Eckart theorem), are outlined in Secs. II and III. Section IV reviews the work of Gould and Paldus[1] and Gould and Battle.[2] This introduces the del operator and summarizes the shift component method leading to the basic segmentation level formulas for the level shifts. The Gould–Battle formula for the ME of the del operator is also reviewed. Section V establishes the Racah factorization lemma for the zero shift ACCS. In Sec. VI we demonstrate that the ACC approach yields exactly the same results as the SCM. This is done first for the basic segmentation level formula and then for the full del-operator MEs. This establishes the complete equivalence of the two approaches and also illustrates the simpler working of the adjoint coupling coefficient approach.

II. BASIC NOTATION AND TERMINOLOGY

In the second quantization approach to molecular orbital theory (i.e., the occupation number representation), we start with a set of $2n$ orthonormal atomic or molecular spin orbitals $\phi^{i\alpha}$ ($i=1,\ldots,n$; $\alpha=0,1$).[1] These span a finite dimensional (f.d.) one-electron space, V.

These one-electron spin–orbitals are constructed by acting upon the vacuum state $|0\rangle$ with the fermion creation operator $X^{\dagger i\mu}$, viz.

$$\phi^{i\mu}=X^{\dagger i\mu}|0\rangle. \tag{11}$$

The corresponding annihilation operator is $X_{i\mu}$. The creation and annihilation operators satisfy the anticommutation relations given by

$$\{X^{\dagger i\mu},X^{\dagger j\nu}\}=\{X_{i\mu},X_{j\nu}\}=0,$$
$$\{X_{i\mu},X^{\dagger j\nu}\}=\delta_i^j\delta_\mu^\nu. \tag{12}$$

The $U(2n)$ generators are formed from them according to

$$E_{j\nu}^{i\mu}=X^{\dagger i\mu}X_{j\nu}. \tag{13}$$

The one-electron state (11) corresponds to an electron in orbital i with spin μ. The N-electron space or Fock space, V_N, is obtained by taking the antisymmetric component of the Nth rank tensor product of V,[3] i.e., $V_N=V^{\otimes N}$. In terms of the occupation number representation a typical state would be denoted by

$$\psi=X^{\dagger i\mu}X^{\dagger j\nu}\cdots X^{\dagger l\alpha}|0\rangle. \tag{14}$$

We see that the anticommutation relations (12) imply that

$$X^{\dagger j\nu}X^{\dagger i\mu}\cdots X^{\dagger l\alpha}|0\rangle=-X^{\dagger i\mu}X^{\dagger j\nu}\cdots X^{\dagger l\alpha}|0\rangle=-\psi. \tag{15}$$

That is, interchanging coordinates reverses the sign of the wave function. In fact, the state (14) corresponds to the Slater determinant wave function in the coordinate representation and the anticommutation relations incorporate the Pauli exclusion principle.[12]

The space V_N of N-particle states gives rise to, that is acts as a carrier space for, an irreducible representation (irrep) of $U(2n)$ with highest weight

$$(1^N,0^{2n-N})=(1,1,\ldots,1,0,\ldots,0), \tag{16}$$

i.e., N ones and $2n-N$ zeros.

As is well known, the Slater determinants are not in general an eigenfunction of spin. Excited states modeled by a single determinant (as in the Hartree–Fock approximation) are characterized by a number of spin states, an effect which is called spin contamination.[12] In addition, a single determinant wave function does not predict the correct dissociation energy for the molecule. Furthermore, because it does not take into account the mutual correlation in the electronic motions, there is a positive energy difference with respect to the exact molecular energy called the correlation energy.

To overcome these defects configuration interaction is employed. In this scheme the wave function is expressed as a sum of configuration state functions (CSFs), viz.

$$\Psi=C_0\psi_0+C_1\psi_1+\cdots+C_N\psi_N. \tag{17}$$

Each CSF (ψ_i) is a linear combination of Slater determinants and is a spin eigenstate. A CSF represents a definite configuration where each electron occupies only one spin–orbital. CI takes into account the problem of electron correlation. The UGA approach to CI affords a highly efficient method of constructing and labeling CSFs.[3]

The appropriate basis to use for spin-independent CI is the Gelfand–Paldus basis. As Paldus first observed in Ref. 3, for the many-electron problem, representations of $U(n)$ have highest weights of the form

$$(2^a,1^b,0^c)=(2,2,\ldots,2,1,\ldots,1,0,\ldots,0). \tag{18}$$

We will denote these representations, which correspond to a two column Young tableau, by $V(a,b,c)$ or $V(p)$ so that $p=(a,b,c)$. Here, the orbital number, n, is given by $n=a+b+c$, the electron number, N, by $N=2a+b$, and the total spin, S, by $S=b/2$.

The Gelfand–Paldus basis for this representation is specified by a labeling scheme obtained from the canonical subgroup chain (2), that is,

$$U(n)\supset U(n-1)\supset U(n-2)\supset\cdots\supset U(2)\supset U(1).$$

Symmetry adaptation to this subgroup chain means that each basis vector is an eigenstate of the Casimir invariants of each subgroup and as a consequence will carry the labels of each subgroup irrep. Since the subgroup chain is canonical, that is, there are no intermediate subgroups, this labeling is complete and specifies the state fully.

A GP basis vector is denoted by

$$|P\rangle=\left|\begin{matrix} a_n & b_n & c_n \\ a_{n-1} & b_{n-1} & c_{n-1} \\ & \cdots & \\ a_2 & b_2 & c_2 \\ a_1 & b_1 & c_1 \end{matrix}\right\rangle=\left|\begin{matrix} p_n \\ p_{n-1} \\ \cdots \\ p_2 \\ p_1 \end{matrix}\right\rangle. \tag{19}$$

Here, P denotes the entire array of Paldus lables, p_i ($i=1,\ldots,n$), and is called the Paldus array. Note that $p=p_n$.

Now, by virtue of Weyl's subgroup branching laws, the irrep $V(p_{m+1})$ of $U(m+1)$ constitutes a reducible representation of $U(m)$ decomposing into at most four irreps $V(p_m)$ of $U(m)$.[3] The Paldus labels p_{m+1} and p_m are related by one of four fundamental shifts Δ_i.[4] That is, $p_{m+1}=p_m+\Delta_i$ ($i=1,\ldots,4$), where Δ_i is one of

$$\Delta_0=(0,0,1);\quad \Delta_1=(0,1,0);$$
$$\Delta_2=(1,-1,1);\quad \Delta_3=(1,0,0). \tag{20}$$

Each GP basis state is a CSF with well-defined spin and occupation number. The number of electrons in the kth orbital of the GP state (19) is given by the eigenvalue of the generator E_{kk}, i.e.,

$$n_k=\sum_{i=1}^{k}p_i^{(k)}-\sum_{i=1}^{k-1}p_i^{(k-1)}=2a_k+b_k-(2a_{k-1}+b_{k-1}). \tag{21}$$

The total number operator $\hat{N}$ is given by

$$\hat{N}=\sum_{\mu=0}^{1}E_\mu^\mu=\sum_{i=1}^{n}E_i^i \tag{22}$$

and has eigenvalue

$$N=\sum_{k=1}^{n} n_k . \tag{23}$$

The orbital $U(n)$ generators, E^i_j $(i,j=1,\ldots,n)$, are obtained from the $U(2n)$ spin–orbital generators by taking a "spin average"

$$E^i_j=\sum_{\mu=0}^{1} E^{i\mu}_{j\nu} . \tag{24}$$

And similarly the spin $U(2)$ generators, E^μ_ν $(\mu,\nu=0,1)$, are given by

$$E^\mu_\nu=\sum_{i=1}^{n} E^{i\mu}_{j\nu} . \tag{25}$$

The orbital $U(n)$ generators satisfy the commutation relations

$$[E^i_j,E^k_l]=\delta^k_j E^i_l-\delta^i_l E^k_j \tag{26}$$

and also the Hermiticity requirement

$$E^{i\dagger}_j=E^j_i . \tag{27}$$

We denote the irrep of $U(2)$ with highest weight $(a+b,a)$ by $V(a+b,a)$.[4–6] The numbers a and b have the same interpretation as for the Paldus label $p=(a,b,c)$ of the irrep $V(p)$. Young diagrams for the irreps $V(a,b,c)$ and $V(a+b,a)$ are conjugate. By analogy with Eq. (19), we will denote the Gelfand–Tsetlin basis state for the irrep $V(a+b,a)$ of $U(2)$ by

$$\left|\begin{matrix} a+b & & a \\ & d & \end{matrix}\right\rangle . \tag{28}$$

In the above, d is an integer which takes the values $a\leqslant d\leqslant a+b$.

For spin-dependent problems it is necessary to obtain the MEs of the $U(2n)$ generators in a basis for the irrep V_N of $U(2n)$ which is symmetry adapted to the spin–orbit subgroup $U(n)\times U(2)$. The resulting spin–orbit basis may be written as

$$\left|\begin{matrix} & (a,b,c) & \\ P' & & d \end{matrix}\right\rangle \equiv \left|\begin{matrix} p \\ P' \end{matrix}\right\rangle \otimes \left|\begin{matrix} a+b & & a \\ & d & \end{matrix}\right\rangle , \tag{29}$$

where P' is the Paldus array for the subgroup $U(n-1)$.

However, since the usual basis of $SU(2)$ denoted by $|SM_s\rangle$, where M_s is the azimuthal spin quantum number, is also an eigenstate of the number operator $\hat{N}$—and hence are also $U(2)$ states[6]—we shall adopt the following notation for the spin–orbit basis states:

$$\left|\begin{matrix} p \\ P_{n-1} \end{matrix}\right\rangle_{M_s} = \left|\begin{matrix} p \\ P_{n-1} \end{matrix}\right\rangle \otimes |SM_s\rangle . \tag{30}$$

Note that the Paldus label p contains all the total spin information required since $S=2a+b$. We will refer to the above as the one-shell spin–orbit basis.

The aim of this series of papers is to find the MEs of the $U(2n)$ generators in a multishell spin–orbit basis. That is, a basis which is symmetry adapted to the subgroup chain (10). Gould *et al.*[8,9] has obtained the MEs of the $U(n)$ generators in a composite Gelfand–Paldus (CGP) basis. The CGP basis is symmetry adapted to the (spin-independent) subgroup chain

$$\begin{aligned} U(n)&=U(n_1+n_2+\cdots+n_r) \\ &\supset U(n_1+n_2+\cdots+n_{r-1})\times U(n_r) \\ &\supset U(n_1+n_2+\cdots+n_{r-2})\times U(n_{r-1})\times U(n_r) \\ &\supset\cdots \\ &\supset U(n_1)\times U(n_2)\times\cdots\times U(n_r). \end{aligned} \tag{31}$$

The CGP states are denoted by

$$\left|\begin{matrix} p_1 & p_2 & & p_r \\ & & \cdots & \\ P'_1 & P'_2 & & P'_r \end{matrix}\right\rangle_{\mathscr{P}} = \left|\begin{matrix} p_1 \\ P'_1 \end{matrix}\right\rangle \otimes \left|\begin{matrix} p_2 \\ P'_2 \end{matrix}\right\rangle \otimes\cdots\otimes \left|\begin{matrix} p_r \\ P'_r \end{matrix}\right\rangle_{\mathscr{P}} . \tag{32}$$

In the above, p_r is the Paldus label for the subgroup $U(n_r)$ and P'_r is the Paldus array for $U(n_r-1)$. The additional Paldus tableau

$$\mathfrak{p}=\begin{bmatrix} \mathfrak{p}_r \\ \mathfrak{p}_{r-1} \\ \cdots \\ \mathfrak{p}_1 \end{bmatrix} \tag{33}$$

tabulates the intermediate coupling labels $\mathfrak{p}_i$, where $\mathfrak{p}_i$ is the Paldus label of the intermediate subgroup $U(n_1+\cdots+n_i)$. Note that $\mathfrak{p}_r=p$, is the Paldus label of the $U(n)$ irrep $V(p)$ and $\mathfrak{p}_1=p_1$. This intermediate tableau contains all the information needed to describe the intermediate coupling scheme suggested by the subgroup chain (31). The subgroup reduction

$$U(n_1+\cdots+n_i)\downarrow U(n_1+\cdots+n_{i-1})\times U(n_i), \quad (i=1,\ldots,r), \tag{34}$$

is multiplicity free for the two-columned irreps of the many-electron problem.

For the spin-dependent multishell problem we adapt the above CGP approach and choose a basis which is symmetry adapted to the (spin-dependent) subgroup chain

$$\begin{aligned} U(2n)&=U(2n_1+2n_2+\cdots+2n_r) \\ &\supset U(2n_1+2n_2+\cdots+2n_{r-1})\times U(2n_r) \\ &\supset\cdots \\ &\supset U(2n_1)\times U(2n_2)\times\cdots\times U(2n_r) \\ &\supset U(n_1)\times U(n_2)\times\cdots\times U(n_r)\times U(2)\times\cdots\times U(2) \\ &\supset U(n_1)\times\cdots\times U(n_r)\times U(2). \end{aligned} \tag{35}$$

This corresponds to

(i) The separation into spin–orbitals of types $1,2,\ldots,r$.
(ii) The separation of spin and orbit.

(iii) The diagonal imbedding $U(2)\times\cdots\times U(2)\supset U(2)$ of angular momentum theory.

The branching rules (BRs) for the reduction of the irrep V_N of $U(2n)$ into irreps of $U(2n_1)\times\cdots\times U(2n_r)$ are given by

$$V_N = \bigoplus_{\substack{N_i\geq 0\\ N_1+\cdots+N_r=N}} V_{N_1}\otimes V_{N_2}\otimes\cdots\otimes V_{N_r}, \tag{36}$$

where $V_{N_i}(i=1,\ldots,r)$ denotes the anti-symmetric tensor representation of $U(2n_i)$ of rank N_i.

In practice, to construct a basis which is symmetry adapted to the subgroup chain (35), we progressively couple the spin–orbit basis states of each shell beginning with shells one and two. As a basis for the spaces $V_{N_1}\otimes V_{N_2}$ we adapt the product spin–orbit basis states

$$\left|\begin{matrix} p_1 \\ P_1' \end{matrix}\right\rangle_{M_1} \otimes \left|\begin{matrix} p_2 \\ P_2' \end{matrix}\right\rangle_{M_2}. \tag{37}$$

That is, the product state having N_1 (respectively, N_2) electrons in shell 1 (respectively, shell 2) with total spin $S_1=b_1/2$ (respectively, $S_2=b_2/2$) and where M_1 (respectively, M_2) is the corresponding spin azimuthal quantum number.

By coupling the spin components of these states we obtain the two-shell spin–orbit states[8]

$$\sum_{M_1,M_2} \langle S_1M_1,S_2M_2|\mathscr{S}_2\mathscr{M}_2\rangle \left|\begin{matrix} p_1 \\ P_1' \end{matrix}\right\rangle_{M_1} \otimes \left|\begin{matrix} p_2 \\ P_2' \end{matrix}\right\rangle_{M_2}$$

$$= \left|\begin{matrix} p_1 & p_2 \\ P_1' & P_2' \end{matrix}\right\rangle_{\mathfrak{p}_2} \otimes |\mathscr{S}_2\mathscr{M}_2\rangle \tag{38}$$

which we denote by

$$\left|\begin{matrix} p_1 & p_2 \\ P_1' & P_2' \end{matrix};\mathfrak{p}_2\right\rangle_{\mathscr{M}_2}. \tag{39}$$

Here, $\mathfrak{p}_2=(a_{12},b_{12},c_{12})$[11] is the intermediate Paldus label for the subgroup $U(n_1+n_2)$, $\mathscr{S}_2=b_{12}/2$ is the intermediate total spin for shells one and two combined, and $\mathscr{M}_2$ is the intermediate total azimuthal spin quantum number, $\mathscr{M}_2=M_1+M_2$. This corresponds to the two-shell spin–orbit state

$$\left|\begin{matrix} p_1 & p_2 \\ P_1' & P_2' \end{matrix}\right\rangle_{\mathfrak{p}_2} \otimes \left|\begin{matrix} a_{12}+b_{12} & & a_{12} \\ & d_{12} & \end{matrix}\right\rangle, \tag{40}$$

where $a_{12}=\frac{1}{2}(N_1+N_2)-\mathscr{S}$, $c_{12}=(n_1+n_2)-a_{12}-b_{12}$, $d_{12}=\mathscr{M}+\frac{1}{2}N_2$.

By progressively coupling the spin–orbit states of shells 1 to r beginning with state (39), we obtain the multishell spin–orbit basis

$$\left|\begin{matrix} p_1 & p_2 & \cdots & p_r \\ P_1' & P_2' & & P_r' \end{matrix};\mathfrak{p}_r\right\rangle_{\mathscr{M}_r} = \left|\begin{matrix} p_1 & p_2 & \cdots & p_r \\ P_1' & P_2' & & P_r' \end{matrix}\right\rangle_{\mathscr{P}} \otimes |SM_s\rangle. \tag{41}$$

As in Eq. (32), $\mathscr{P}$ is the intermediate Paldus array and $\mathscr{M}_r$ is now the total azimuthal component of total spin S. The spin coupling scheme

$$[\cdots[[[S_1\otimes S_2]^{\mathscr{S}_2}\otimes S_3]^{\mathscr{S}_3}\otimes\cdots\otimes S_{r-1}]^{\mathscr{S}_{r-1}}\otimes S_r]^{\mathscr{S}_r=S} \tag{42}$$

corresponds to the usual $L-S$ coupling scheme of atomic calculations.

We now review the basic theory of tensor operators and the Wigner–Eckart theorem. This enables us to introduce frequently used terms (and the corresponding notation) in the context in which they are developed. They are an essential ingredient in what follows.

III. TENSOR OPERATORS AND THE WIGNER–ECKART THEOREM

A tensor operator under some group, G say, is a set of linearly independent operators indexed like the basis vectors of the carrier space of a representation of G. Under the operations of G the operator set transforms in the same way as the basis set.

More formally, for group G let V with basis $v_1,\ldots,v_k$ so that $\dim V=k$, be a carrier space for a representation π of G. That is, if $x\in L$ the Lie algebra of G, we have $\pi(x)\in \mathrm{End}(V)$. The collection of operators $\{T_i\}_{k=1}^k$ is called a tensor operator of G if the components T_i transform according to

$$[x,T_i]=\sum_{j=1}^{n} \pi(x)_i^j T_j. \tag{43}$$

Here, $\pi(x)_i^j$ is the matrix representing $x\in L$ in the basis $\{v_i\}_{i=1}^k$.

We shall follow Gould and Chandler[5] and refer to $\{T_i\}_{i=1}^k$ as a tensor operator of rank π. For example, if $V(\lambda)$ is an irrep of $U(n)$ we say that T is an irreducible tensor operator of rank λ.

The concept of a vector operator is a particular case of the general definition (43). The indexed set $\{\psi_i\}_{i=1}^n$ is a vector operator of $U(n)$ if,[4]

$$[E_j^i,\psi_k]=\delta_{kj}\psi_i. \tag{44}$$

This follows from the fact that if V is the fundamental vector representation of $U(n)$ with basis $\{e_i\}_{i=1}^n$ and highest weight of the form $(1,\dot{0})$, then the action of the $U(n)$ generators is given by

$$E_j^i e_k=\delta_{kj}e_i. \tag{45}$$

Similarly, if V^* is the contragredient vector representation of $U(n)$ with basis $\{\bar{e}_i\}_{i=1}^n$ and highest weight $(\dot{0},-1)$, then the action of the $U(n)$ generators is given by

$$E^i_j \bar{e}_k = -\delta_{kj}\bar{e}_i . \tag{46}$$

This yields the transformation law for a contragredient vector operator, ϕ_k say,

$$[E^i_j,\phi_k] = -\delta_{ik}\phi_j . \tag{47}$$

Note that the Hermitian conjugate of a $U(n)$ vector operator $\psi_i^\dagger = (\psi_i)^\dagger$ is a $U(n)$ contragredient vector operator.

The action of the $U(n)$ generators E^i_j $(i,j=1,\ldots,n)$ on a GP basis state $\left|{p \atop P}\right\rangle$ may be determined by examining their properties as a vector operator. If $U(m+1)$ and $U(m)$ are two subgroups occurring in the canonical chain (2) then the $U(m+1)$ generators $E_{i,m+1}$ (respectively, $E_{m+1,i}$) $(i,j=1,\ldots,m)$ constitute a vector (respectively, contragredient vector) operator of $U(m)$.[4]

From Gould and Chandler,[4] the action of a $U(n)$ vector operator $\psi_i : \mathscr{H} \to \mathscr{H}$ on an irrep $V(p)$ of $U(n)$ contained in a Hilbert space $\mathscr{H}$, is equivalent to investigating the action of an intertwining operator ψ defined by the mapping

$$\psi: V\otimes\mathscr{H} \to \mathscr{H}. \tag{48}$$

Here, we assume also that the Hilbert space $\mathscr{H}$ is the multiplicity free direct sum of all the two-column irreps of $U(n)$ pertinent to the many-electron problem. It is also explicitly understood that $\mathscr{H}$ is restricted to the subspace $V(p)$. An intertwining operator is one which commutes with the action of the generators, viz.

$$E^i_j\psi = \psi[\pi(E^i_j)\otimes\mathbf{1} + \mathbf{1}\otimes\pi_p(E_{ij})]. \tag{49}$$

$\pi(E_{ij})$ and $\pi_p(E_{ij})$ are the operators representing the generator E_{ij} on the spaces V and $V(p)$, respectively, and E_{ij} is understood to act on $\mathscr{H}$.

In terms of components

$$\psi_i v = \psi(e_i\otimes v), \quad v\in V(p)\subseteq\mathscr{H}. \tag{50}$$

The product space decomposes into irreps of $U(n)$ via

$$V\otimes V(p) = \bigoplus_{\alpha=1}^{3} V(p'_\alpha), \tag{51}$$

where the Paldus label p'_α corresponds to one of the highest weights

$$\begin{aligned} p'_1 &\equiv (2^a,1^b,1,0^{c-1}),\\ p'_2 &\equiv (2^a,2,1^{b-1},0^c),\\ p'_3 &\equiv (3,2^{a-1},1^b,0^c). \end{aligned} \tag{52}$$

The third is nonlexical for the two-column irreps of the many-electron problem and does not contribute.

Now, the effect of a vector operator on a GP basis state is to shift the entries of the Paldus labels in some of the levels of the array. The vector operator may be decomposed into a sum of independent shift components which are found by the use of projection operators.[4]

We will denote the projection operator of $V\otimes V(p)$ onto the space $V(p'_\alpha)$ by $\bar{P}[\alpha]$. This can then be used to project out the shift components of a vector operator ψ_i, viz.

$$\psi_i = \sum_{\alpha=1}^{3} \psi[\alpha]_i . \tag{53}$$

Since ψ is an intertwining operator

$$\psi V(p'_\alpha) \subseteq V(p'_\alpha) \tag{54}$$

from which it follows that the operator

$$\psi[\alpha] = \psi\bar{P}[\alpha] \tag{55}$$

is an intertwining operator from $V\otimes V(p)$ onto $V(p'_\alpha) \leqslant \mathscr{H}$. In component form

$$\psi[\alpha]_i = \psi_j \bar{P}[\alpha]_{ji} \tag{56}$$

so that

$$\psi[\alpha]_i v = \psi\bar{P}[\alpha](e_i\otimes v) \in V(p'_\alpha). \tag{57}$$

That is, the shift component $\psi[\alpha]_i$ changes the representation labels of $U(n)$ from p to p'_1 or p'_2. We can write these shifts as

$$p'_1 = p+\delta_1, \quad p'_2 = p+\delta_2, \tag{58}$$

The increments

$$\delta_1 = (0,1,-1) \quad \text{and} \quad \delta_2 = (1,-1,0) \tag{59}$$

are called the fundamental vector shifts.

The WE theorem enables the ME of a vector or tensor operator to be expressed as the product of a scalar, depending only on the irrep label, and a coupling coefficient (or as a sum of such products).

From Eqs. (53) and (57) we see that the only nonzero MEs of ψ_i are given by

$$\left\langle {p'_\alpha \atop P'} \middle| \psi_i \middle| {p \atop p} \right\rangle = \left\langle {p'_\alpha \atop P'} \middle| \psi[\alpha]_i \middle| {p \atop P} \right\rangle. \tag{60}$$

$\left|{p'_\alpha \atop P'}\right\rangle$ and $\left|{p \atop P}\right\rangle$ are GP states belonging to $V(p'_\alpha)$, $V(p)\subseteq\mathscr{H}$, respectively. Now, using the fact that $\psi[\alpha] = \psi\bar{P}[\alpha]$ [Eq. (56)] we may write

$$\begin{aligned} \left\langle {p'_\alpha \atop P'} \middle| \psi_i \middle| {p \atop P} \right\rangle &= \left\langle {p'_\alpha \atop P'} \middle| \psi\bar{P}[\alpha] \middle| e_i \otimes {p \atop P} \right\rangle \\ &= \sum_{P''} \left\langle {p'_\alpha \atop P'} \middle| \psi \middle| {p'_\alpha \atop P''} \right\rangle \left\langle {p'_\alpha \atop P''} \middle| e_i ; {p \atop P} \right\rangle, \end{aligned} \tag{61}$$

where the sum is over all Paldus tableau for the subspace $V(p'_\alpha) \subseteq V \otimes V(p)$ and where also, we have used the fact that

$$P[\alpha] = \sum_{P''} \left| {p'_\alpha \atop P''} \right\rangle \left\langle {p'_\alpha \atop P''} \right|.$$

On the subspace $V\otimes V(p)$ we may define an intertwining operator $\tilde{\psi}$ by

$$\tilde{\psi}: V(p'_\alpha) \to V(p'_\alpha)$$

such that

$$\left| {p'_\alpha \atop P''} \right\rangle \mapsto \sum_{P'} \left| {p'_\alpha \atop P'} \right\rangle \left\langle {p'_\alpha \atop P'} \middle| \psi \middle| {p'_\alpha \atop P''} \right\rangle. \tag{62}$$

This result follows from the intertwining property of ψ.

Now, if we apply Schur's lemma[15,16] we see that this map must reduce to a scalar multiple, R_α, of the identity on $V(p'_\alpha)\subseteq V\otimes V(p)$. That is,

$$\left\langle \begin{matrix} p'_\alpha \\ P' \end{matrix} \middle| \psi \middle| \begin{matrix} p'_\alpha \\ P'' \end{matrix} \right\rangle = R_\alpha \cdot \delta_{P''P'}. \tag{63}$$

Thus Eq. (61) becomes

$$\left\langle \begin{matrix} p'_\alpha \\ P' \end{matrix} \middle| \psi_i \middle| \begin{matrix} p \\ P \end{matrix} \right\rangle = R_\alpha \cdot \left\langle \begin{matrix} p'_\alpha \\ P' \end{matrix} \middle| e_i ; \begin{matrix} p \\ P \end{matrix} \right\rangle. \tag{64}$$

This is the Wigner–Eckart theorem for $U(n)$ vector operators. It expresses the matrix element of the vector operator as the product of a reduced matrix element (RME), R_α, which depends only on the $U(n)$ irrep labels p, p_α and a vector coupling coefficient (VCC) or Wigner coefficient $\langle {}^{p_\alpha}_{P'} | e_i ; {}^{p}_{P} \rangle$. The RME is also written

$$R_\alpha = \langle p_\alpha \| \psi \| p \rangle. \tag{65}$$

Similarly, for the contragredient vector operator $\psi_i^\dagger$ we may equate its action to that of an intertwining operator $\psi^\dagger$ by the mapping

$$\psi^\dagger : V^* \otimes \mathscr{H} \to \mathscr{H}, \tag{66}$$

and the component definition

$$\psi_i^\dagger v = \psi^\dagger (\bar{e}_i \otimes v), \quad v \in V(p). \tag{67}$$

The product space $V^*\otimes V(p)$ has a similar expansion

$$V^* \otimes V(p) = \bigoplus_{\alpha=1}^{3} V(p'_\alpha), \tag{68}$$

where p'_α is now one of

$$\begin{aligned} p'_1 &= (2^a, 1^{b-1}, 0, 0^c), \\ p'_2 &= (2^{a-1}, 1, 1^b, 0^c), \\ p'_3 &= (2^a, 2^b, 0^{c-1}, -1). \end{aligned} \tag{69}$$

Again p'_3 is nonlexical. By an exactly similar development to that of the above, the shift components of $\psi^\dagger$ are given by

$$\psi_i^\dagger = \sum_{\alpha=1}^{3} \psi^\dagger[\alpha]_i \tag{70}$$

and with the use of the projector $P[\alpha]$, which projects $V^*\otimes V(p)$ onto $V(p'_\alpha)$, they are given by

$$\psi^\dagger[\alpha]_i = \psi_j^\dagger P[\alpha]_{ji}. \tag{71}$$

Schurs' lemma now yields the WE theorem for contragredient vector operators.

$$\left\langle \begin{matrix} p'_\beta \\ P'' \end{matrix} \middle| \psi_i^\dagger \middle| \begin{matrix} p \\ P' \end{matrix} \right\rangle = \bar{R}_\beta \cdot \left\langle \begin{matrix} p'_\beta \\ P'' \end{matrix} \middle| \bar{e}_i ; \begin{matrix} p \\ P' \end{matrix} \right\rangle. \tag{72}$$

By projecting out the shift components of the $U(n)$ generators and applying the WE theorem, Gould and Chandler[4] were able to write down formulas for the $U(n)$ generator matrix elements. The MEs of the elementary generators are denoted by

$$N_m^r = \left\langle \begin{matrix} p_{m+1} \\ p_m + \delta_r \\ P_{m-1} \end{matrix} \middle| E^m_{m+1} \middle| \begin{matrix} p_{m+1} \\ p_m \\ P_{m-1} \end{matrix} \right\rangle \tag{73}$$

and

$$\bar{N}_m^r = \left\langle \begin{matrix} p_{m+1} \\ p_m - \delta_r \\ P_{m-1} \end{matrix} \middle| E^{m+1}_{m} \middle| \begin{matrix} p_{m+1} \\ p_m \\ P_{m-1} \end{matrix} \right\rangle. \tag{74}$$

The formulas for N_m^r and $\bar{N}_m^r$ may be found in Refs. 4 and 7. Here (and in what follows), the Paldus labels from p_n to the top label in Eqs. (73) and (74) have been suppressed for ease of presentation since they are unshifted. P_{m-1} is an allowed Paldus tableau for the subgroup $U(m-1)$.

The nonelementary generator MEs are similarly denoted by

$$N\begin{pmatrix} m+p-1 & & m \\ & \cdots & \\ i_{m+p-1} & & i_m \end{pmatrix} = \left\langle \begin{matrix} p_{m+p} \\ p_{m+p-1} + \delta_{i_{m+p-1}} \\ \vdots \\ p_m + \delta_{i_m} \\ P_{m-1} \end{matrix} \middle| E^m_{m+p} \middle| \begin{matrix} p_{m+p} \\ p_{m+p-1} \\ \vdots \\ p_m \\ P_{m-1} \end{matrix} \right\rangle \tag{75}$$

and

$$\bar{N}\begin{pmatrix} m+p-1 & & m \\ & \cdots & \\ i_{m+p-1} & & i_m \end{pmatrix} = \left\langle \begin{matrix} p_{m+p} \\ p_{m+p-1} - \delta_{i_{m+p-1}} \\ \vdots \\ p_m - \delta_{i_m} \\ P_{m-1} \end{matrix} \middle| E^{m+p}_{m} \middle| \begin{matrix} p_{m+p} \\ p_{m+p-1} \\ \vdots \\ p_m \\ P_{m-1} \end{matrix} \right\rangle. \tag{76}$$

Explicit formulas for them are also given in Refs. 4 and 7.

Also, following the notation of Gould and Chandler in the above, we shall denote the general form of the VCC by

$$V\begin{pmatrix} n & & m \\ & \cdots & \\ i_n & & i_m \end{pmatrix} = \left\langle \begin{matrix} p_n + \delta_{i_n} \\ \vdots \\ p_m + \delta_{i_m} \\ P_{m-1} \end{matrix} \middle| e_m ; \begin{matrix} p_n \\ \vdots \\ p_m \\ P_{m-1} \end{matrix} \right\rangle \tag{77}$$

and that of the CVCC by

$$\bar{V}\begin{pmatrix} n & & m \\ & \cdots & \\ i_n & & i_m \end{pmatrix} = \left\langle \begin{matrix} p_n - \delta_{i_n} \\ \vdots \\ p_m - \delta_{i_m} \\ P_{m-1} \end{matrix} \middle| \bar{e}_m ; \begin{matrix} p_n \\ \vdots \\ p_m \\ P_{m-1} \end{matrix} \right\rangle. \tag{78}$$

With this notation we may apply the WE theorem to obtain the MEs of the $U(n)$ generators E^m_{m+p}, viz.

$$N\begin{pmatrix} m+p-1 & & m \\ & \cdots & \\ i_{m+p-1} & & i_m \end{pmatrix} = [R^{i_{m+p-1}}_{m+p-1}]^{1/2}\cdot V\begin{pmatrix} m+p-1 & & m \\ & \cdots & \\ i_{m+p-1} & & i_m \end{pmatrix} \tag{79}$$

which is shorthand for

$$\left\langle \begin{matrix} p_{m+p} \\ p_{m+p-1}+\delta_{i_{m+p-1}} \\ \vdots \\ p_m+\delta_{i_m} \\ P_{m-1} \end{matrix} \middle| E^m_{m+p} \middle| \begin{matrix} p_{m+p} \\ p_{m+p-1} \\ \vdots \\ p_m \\ P_{m-1} \end{matrix} \right\rangle = \left\langle \begin{matrix} p_{m+p} \\ p_{m+p-1}+\delta_{i_{m+p-1}} \end{matrix} \middle\| E \middle\| \begin{matrix} p_{m+p} \\ p_{m+p-1} \end{matrix} \right\rangle \times \left\langle \begin{matrix} p_{m+p-1}+\delta_{i_{m+p-1}} \\ \vdots \\ p_m+\delta_{i_m} \\ P_{m-1} \end{matrix} \middle| e_m; \begin{matrix} p_{m+p-1} \\ \vdots \\ p_m \\ P_{m-1} \end{matrix} \right\rangle . \tag{80}$$

Formulas for the squares of the RMEs, denoted by $R^{i_n}_n$, are given in Refs. 4 and 7 and formulas for the VCCs and CVCCs are found in Refs. 5 and 7. The MEs of the raising generators E^{m+p}_m are obtained similarly, viz.

$$\bar{N}\begin{pmatrix} m+p-1 & & m \\ & \cdots & \\ i_{m+p-1} & & i_m \end{pmatrix} = [\bar{R}^{i_{m+p-1}}_{m+p-1}]^{1/2}\cdot \bar{V}\begin{pmatrix} m+p-1 & & \\ & \cdots & \\ i_{m+p-1} & & i_m \end{pmatrix}. \tag{81}$$

To determine the MEs of the $U(2n)$ generators, $E^{i\mu}_{j\nu}$, we set out (initially) to follow a similar development to that of the above. Here, the aim is to calculate the MEs with respect to the SO basis (30). The basic motivation is that the $U(2n)$ generators transform as the representation $\mathrm{Adj}[U(n)]\otimes\mathrm{Adj}[U(2)]$.[5] That is, they transform as an adjoint tensor operator of both $U(n)$ and $U(2)$.

Following Gould and Paldus,[1] we note that it is a general property of $U(2)$ ATOs that they may be resolved into spin-shift components

$$E^{i\mu}_{j\nu}=E(-)^{i\mu}_{j\nu}+E(0)^{i\mu}_{i\nu}+E(+)^{i\mu}_{j\nu}. \tag{82}$$

The components $E(\pm)^{i\mu}_{j\nu}$ increase (respectively, decrease) the total spin S of the state (30) by one unit. The zero-shift component $E(0)^{i\mu}_{j\nu}$ has no effect on the spin.

Using the method of polynomial identities (Bracken and Green[17] and Gould[18]) to construct $U(2)$ projection operators $P[1]^\mu_\nu$ and $P[2]^\mu_\nu$ (μ,ν=0,1), we can write the shift components of Eq. (82) as

$$E(+)^{i\mu}_{j\nu}=P[1]^\mu_\tau E^{i\tau}_{j\sigma}P[2]^\sigma_\nu,$$

$$E(-)^{i\mu}_{j\nu}=P[2]^\mu_\tau E^{i\tau}_{j\sigma}P[1]^\sigma_\nu, \tag{83}$$

$$E(0)^{i\mu}_{j\nu}=P[1]^\mu_\tau E^{i\tau}_{j\sigma}P[1]^\sigma_\nu+P[2]^\mu_\tau E^{i\tau}_{j\sigma}P[2]^\sigma_\nu.$$

These shift components alter the Paldus labels $p=(a,b,c)$ of the group $U(n)$ according to[5]

$$E(\pm):p\mapsto p+\epsilon_\pm, \quad E(0):p\mapsto p+\epsilon_0, \tag{84}$$

where ϵ_α (α=0,$\pm$) denotes the adjoint shifts

$$\epsilon_+=\delta_1-\delta_2=(-1,+2,-1)=+\epsilon,$$
$$\epsilon_-=\delta_2-\delta_1=(+1,-2,+1)=-\epsilon, \tag{85}$$
$$\epsilon_0=(0,0,0).$$

To obtain the MEs of the nonzero spin-shift operators we apply the $U(n)\times U(2)$ WE theorem to obtain

$${}_{M'_s}\left\langle \begin{matrix} p\pm\epsilon \\ P' \end{matrix} \middle| E(\pm)^{i\mu}_{j\nu} \middle| \begin{matrix} p \\ P \end{matrix} \right\rangle_{M_s} = \langle p\pm\epsilon\|E\|p\rangle\cdot\left\langle \begin{matrix} p\pm\epsilon \\ P \end{matrix} \middle| e_i\otimes\bar{e}_j; \begin{matrix} p \\ P \end{matrix} \right\rangle \times\langle SM'_s|e_\mu\otimes\bar{e}_\nu;SM_s\rangle. \tag{86}$$

The first term in Eq. (86) is the $U(2n):U(n)\times U(2)$ RME and the second and third terms are a $U(n)$ and an $SU(2)$ adjoint coupling coefficient, respectively. We recall that $\{e_i\}$ (respectively, $\{\bar{e}_i\}$) denotes the usual basis for the fundamental vector (respectively, contragredient vector) representation, of $U(n)$. $\{e_i\otimes\bar{e}_j\}$ is the GT basis for the adjoint representation.[5] The ME (86) vanishes unless

$$M'_s=M_s+\nu-\mu \quad \text{and } S'=S\pm1, \tag{87}$$

a result which follows from the form of the adjoint shifts (84).

However, the WE theorem (in its usual form) cannot be applied to the zero-shift components $E(0)^{i\mu}_{j\nu}$.[1] This follows because of the multiplicities implied by the fact that there is more than one zero-shift ATO for $U(n)$ or $U(2)$.

Now, from Louck and Biedenharm,[19] we know that the set of all tensor operators on an irrep $V(\lambda)$, say, is itself a vector space with the same dimensionality, $D[\lambda]$, as the irrep. So that each tensor operator has $D[\lambda]$ independent components. That is, we can find a basis for the operator space.

The operator space can be partitioned into classes according to the nature of the shift action of the tensor operator. The number of independent operators of a particular shift type is called the multiplicity. In order to apply the WE in its usual form the multiplicity must be one, as is the case for the nonzero shift components $E(\pm)^{i\mu}_{j\nu}$.

For $U(n)$ there are three independent zero-shift operators, viz. δ^i_j, E^i_j, and $(E^2)^i_j$ and correspondingly there are two independent zero-shift $U(2)$ ATOs, namely, δ^μ_ν and E^μ_ν. For the MEs of the zero-shift components, $E(0)^{i\mu}_{j\nu}$, Gould and Chandler[6,7] have shown that the WE theorem can be applied in a special form. However, this approach leads to complex formulas and a formalism that is difficult to implement on the computer.

We will now review the del operator formalism of Gould and Paldus.[1] The results obtained in their work constitute the starting point for our alternative derivation of the $U(2n)$ generator MEs via the use of adjoint coupling coefficients and the Racah factorization lemma.

IV. THE DEL OPERATOR

The del operator, defined in Eq. (7) by

$$\Delta(n)^i_j = E(E+N/2-n-2)^i_j = E(E+\gamma)^i_j \tag{88}$$

was introduced by Gould and Paldus[1] in the context of determining the MEs of the zero-shift components of the $U(2n)$ generators. $\Delta(n)^i_j$ is an ATO of $U(n)$ and as such does not shift the representation labels of $U(n)$.

Of central importance in this work is the unusual vanishing property of this operator. If $|v\rangle = |{}^p_P\rangle$ is a $U(n)$ GP state with level i doubly occupied or level j unoccupied, then the action of $\Delta(n)^i_j$ on this state is given by

$$\Delta(n)^i_j|v\rangle = 0. \tag{89}$$

Indeed, for given n, $\Delta(n)^i_j$ is the unique, up to a multiple, ATO with this property. A lemma proving this is given in Appendix B. Moreover[14] $\Delta(n)^i_j$ vanishes on any irrep on $U(n)$ with total spin $S=\frac{1}{2}b=0$.

Gould and Paldus have shown in Ref. 1 that the MEs of the zero-shift components of the $U(2n)$ generators in the SO basis may be expressed in terms of the del operator. They are given by

$${}_{M_s'=0}\left\langle \begin{matrix} p \\ P' \end{matrix} \middle| E(0)^{i\mu}_{j\nu} \middle| \begin{matrix} p \\ P \end{matrix} \right\rangle_{M_s=0} = \frac{1}{2}\,\delta^\mu_\nu \left\langle \begin{matrix} p \\ P' \end{matrix} \middle| E^i_j \middle| \begin{matrix} p \\ P \end{matrix} \right\rangle, \quad S=0,$$

and

$$\begin{aligned} {}_{M_s'}\left\langle \begin{matrix} p \\ P' \end{matrix} \middle| E(0)^{i\mu}_{j\nu} \middle| \begin{matrix} p \\ P \end{matrix} \right\rangle_{M_s} &= \frac{1}{2}\,\delta^\mu_\nu \delta_{M_s',M_s} \left\langle \begin{matrix} p \\ P' \end{matrix} \middle| E^i_j \middle| \begin{matrix} p \\ P \end{matrix} \right\rangle \\ &\quad + \frac{-1}{2S(S+1)} \left\langle \begin{matrix} p \\ P' \end{matrix} \middle| \Delta^i_j \middle| \begin{matrix} p \\ P \end{matrix} \right\rangle \\ &\quad \times \langle SM_s'|\tilde{E}^\mu_\nu|SM_s\rangle, \quad S\neq 0. \end{aligned} \tag{90}$$

In the above $\tilde{E}^\mu_\nu$ denotes the $SU(2)$ generators which are given by

$$\tilde{E}^\mu_\nu = E^\mu_\nu - N/2\,\delta^\mu_\nu. \tag{91}$$

The MEs of the nonzero shift components may be evaluated by direct application of the WE theorem, as indicated by Eq. (86). However, Gould and Paldus in Ref. 1 have shown that these also may be expressed in terms of the del operator. By the use of the identity

$$\begin{aligned} \Delta(n+1)^i_j \equiv \hat{\Delta}^i_j &= E^i_{n+1}E^{n+1}_j + \Delta(n)^i_j \\ &\quad + (\tfrac{1}{2}E^{n+1}_{n+1} - 1)E^i_j, \quad 1\leqslant i,j\leqslant n, \end{aligned} \tag{92}$$

the RMEs of Eq. (86) may be evaluated and the adjoint coupling coefficients may be related to the MEs of the del operator. We refer to Eq. (92) later in this paper and a proof of it is given in Appendix B.

The MEs of the nonzero shift components of the $U(2n)$ generators are then given by

$$\begin{aligned} {}_{M_s'}\left\langle \begin{matrix} p+\epsilon \\ P' \end{matrix} \middle| E(+)^{i\mu}_{j\nu} \middle| \begin{matrix} p \\ P \end{matrix} \right\rangle_{M_s} &= (-1)^b \left(\frac{b+2}{b+3}\right)^{1/2} \\ &\quad \times \left\langle \begin{matrix} p+(0,1,0) \\ p+\epsilon \\ P' \end{matrix} \middle| \hat{\Delta}^i_j \middle| \begin{matrix} p+(010) \\ p \\ P \end{matrix} \right\rangle \\ &\quad \times \langle S+1, M_s'|e_\mu \otimes \bar{e}_\nu; SM_s\rangle \end{aligned}$$

and

$$\begin{aligned} &{}_{M_s'}\left\langle \begin{matrix} p-\epsilon \\ P' \end{matrix} \middle| E(-)^{i\mu}_{j\nu} \middle| \begin{matrix} p \\ P \end{matrix} \right\rangle_{M_s} \\ &= (-1)^b \left(\frac{b}{b-1}\right)^{1/2} \\ &\quad \times \left\langle \begin{matrix} p+(1,-1,1) \\ p-\epsilon \\ P' \end{matrix} \middle| \hat{\Delta}^i_j \middle| \begin{matrix} p+(1,-1,1) \\ p \\ P \end{matrix} \right\rangle \\ &\quad \times \langle S-1, M_s'|e_\mu \otimes \bar{e}_\nu; SM_s\rangle. \end{aligned} \tag{93}$$

The formulas of Eqs. (90) and (93) now permit a unified approach to the determination of the MEs of the $U(2n)$ generators. It is computationally more convenient to use the del-operator formulas for the nonzero shift components as they are also required for the zero-shift case.

The MEs of the $U(n)$ generators, E^i_j, were obtained in Refs. 4 and 7 and formulas for the adjoint coupling coefficients are given in Refs. 5 and 7. It remains then to obtain the MEs of the del operator. Gould and Battle[2] have derived formulas for the del-operator MEs in a GP basis. The method used by them is analogous to the shift component method of Gould and Chandler,[4,7] which was used previously to evaluate the MEs of the $U(n)$ generators. In the outline that follows we have summarized the results of Gould and Battle.[2] Later in this paper we will reproduce these same formulas by the use of an alternative method which relies on adjoint coupling coefficients.

The $U(n+1)$ ATO $\Delta(n+1)^i_j$ may be decomposed into $U(n)$ shift components

$$\Delta(n+1)_{ij} = \sum_{\alpha_n} \Delta\begin{pmatrix} n \\ \alpha_n \end{pmatrix}_{ij}, \tag{94}$$

where for notational convenience we have written $E_{ij}\equiv E^i_j$ and where also

$$\Delta\begin{pmatrix} n \\ \alpha_n \end{pmatrix}_{ij} : p_n \to p_n + \epsilon_{\alpha_n}, \quad (\alpha_n = +,0,-), \tag{95}$$

is the $U(n)$ adjoint shift component of $\Delta(n+1)_{ij}$.

By exploiting the unique vanishings property of the del operator, it is found that

$$\Delta\begin{pmatrix} n \\ 0 \end{pmatrix}_{ij} = \frac{1+b_{n+1}+\Delta b_{n+1}}{1+b_{n+1}} \cdot \Delta(n)_{ij}$$

$$= (\alpha_n + 1)\cdot \Delta(n)_{ij}, \tag{96}$$

which defines

$$\alpha_n = \frac{\Delta b_{n+1}}{1+b_{n+1}}, \quad \Delta b_{n+1} = b_{n+1} - b_n. \tag{97}$$

Note that used as a subscript, $\alpha_n = +,0,-$.

Similarly the $U(n-1)$ zero-shift component is found to be

$$\Delta\begin{pmatrix} n & n-1 \\ \pm & 0 \end{pmatrix}_{ij} = \Delta\begin{pmatrix} n \\ \pm \end{pmatrix}_{nn} \cdot \frac{2}{b_{n-1}(b_{n-1}+2)} \cdot \Delta(n-1)_{ij}. \tag{98}$$

Equations (96) and (98), are used to obtain recursive formulas for the basic shift components of the diagonal del operators. For the operator $\Delta(n+1)_{n-1,n-1}$ we have the decomposition

$$\Delta(n+1)_{n-1,n-1} = \sum_{\alpha_n} \sum_{\alpha_{n-1}} \Delta\begin{pmatrix} n & n-1 \\ \alpha_n & \alpha_{n-1} \end{pmatrix}_{n-1,n-1}. \tag{99}$$

Noting that the shift components $\Delta(\begin{smallmatrix} n & n-1 \\ + & - \end{smallmatrix})$ and $\Delta(\begin{smallmatrix} n & n-1 \\ - & + \end{smallmatrix})$ are nonlexical, we see that there are at most nine (9) final states.[2] The allowed shift components are given by

$$\Delta\begin{pmatrix} n & n-1 \\ 0 & 0 \end{pmatrix}_{n-1,n-1} = (1+\alpha_n)(1+\alpha_{n-1})\Delta(n-1)_{n-1,n-1}, \tag{100a}$$

$$\Delta\begin{pmatrix} n & n-1 \\ \pm & 0 \end{pmatrix}_{n-1,n-1} = \frac{2}{b_{n-1}(2+b_{n-1})} \cdot \Delta\begin{pmatrix} n \\ \pm \end{pmatrix}_{nn} \cdot \Delta(n-1)_{n-1,n-1}, \tag{100b}$$

$$\Delta\begin{pmatrix} n & n-1 \\ 0 & \pm \end{pmatrix}_{n-1,n-1} = (1+\alpha_n)\cdot \Delta\begin{pmatrix} n-1 \\ \pm \end{pmatrix}_{n-1,n-1}, \tag{100c}$$

$$\Delta\begin{pmatrix} n & n-1 \\ \pm & \pm \end{pmatrix}_{n-1,n-1} = X\begin{pmatrix} n & n-1 \\ \pm & \pm \end{pmatrix}_{n-1,n-1}. \tag{100d}$$

X_{ij} is the two-body operator $E_{in+1}E_{n+1j}$.[2,5]

When the MEs of these basic shift components are evaluated, it is found that the resulting expressions may be factorized into a product of distinct segments.[2] These are analogous to the segment level form of the MEs of the $U(n)$ generators which were obtained by Shavitt.[20]

They are

$$\left\langle \begin{matrix} p_{n+1} \\ p_n \\ p_{n-1} \\ P_{n-2} \end{matrix} \middle| \Delta\begin{pmatrix} n & n-1 \\ 0 & 0 \end{pmatrix}_{n-1,n-1} \middle| \begin{matrix} p_{n+1} \\ p_n \\ p_{n-1} \\ P_{n-2} \end{matrix} \right\rangle = T^{n+1}(0)\cdot T^n(0,0)\cdot T_{n-1}(0),$$

$$\left\langle \begin{matrix} p_{n+1} \\ p_n \pm \epsilon \\ p_{n-1} \\ P_{n-2} \end{matrix} \middle| \Delta\begin{pmatrix} n & n-1 \\ \pm & 0 \end{pmatrix}_{n-1,n-1} \middle| \begin{matrix} p_{n+1} \\ p_n \\ p_{n-1} \\ P_{n-2} \end{matrix} \right\rangle = T^{n+1}(\pm)\cdot T^n(\pm,0)\cdot T_{n-1}(0),$$

$$\tag{101}$$

$$\left\langle \begin{matrix} p_{n+1} \\ p_n \\ p_{n-1} \pm \epsilon \\ P_{n-2} \end{matrix} \middle| \Delta\begin{pmatrix} n & n-1 \\ 0 & \pm \end{pmatrix}_{n-1,n-1} \middle| \begin{matrix} p_{n+1} \\ p_n \\ p_{n-1} \\ P_{n-2} \end{matrix} \right\rangle = T^{n+1}(0)\cdot T^n(0,\pm)\cdot T_{n-1}(\pm),$$

$$\left\langle \begin{matrix} p_{n+1} \\ p_n \pm \epsilon \\ p_{n-1} \pm \epsilon \\ P_{n-2} \end{matrix} \middle| \Delta\begin{pmatrix} n & n-1 \\ \pm & \pm \end{pmatrix}_{n-1,n-1} \middle| \begin{matrix} p_{n+1} \\ p_n \\ p_{n-1} \\ P_{n-2} \end{matrix} \right\rangle = T^{n+1}(\pm)\cdot T^n(\pm,\pm)\cdot T_{n-1}(\pm).$$

The segments $T^{n+1}(\alpha_n)$, $T^n(\alpha_n,\alpha_{n-1})$ and $T_n(\alpha_n)$ are listed in Appendix C.

The MEs of the general diagonal operator $\Delta(n+1)_{mm}$ are obtained by continuing this recursive reduction. They are given by

$$\left\langle \begin{matrix} p_{n+1} \\ p_n+\epsilon_{\alpha_n} \\ p_{n-1}+\epsilon_{\alpha_{n-1}} \\ \vdots \\ p_m+\epsilon_{\alpha_m} \\ P_{m-1} \end{matrix} \right| \Delta(n+1)_{mm} \left| \begin{matrix} p_{n+1} \\ p_n \\ p_{n-1} \\ \vdots \\ p_m \\ P_{m-1} \end{matrix} \right\rangle = T^{n+1}(\alpha_n)\cdot \prod_{r=m+1}^{n} T^r(\alpha_r,\alpha_{r-1})\cdot T_m(\alpha_m) = \Delta^* \begin{pmatrix} n & & m+1 \\ & \cdots & \\ \alpha_n & & \alpha_{m+1} \end{pmatrix} \cdot T_m(\alpha_m), \tag{102}$$

where it is convenient to define

$$\Delta^* \begin{pmatrix} n & & m+1 \\ & \cdots & \\ \alpha_n & & \alpha_{m+1} \end{pmatrix} = T^{n+1}(\alpha_m)\cdot \prod_{r=m+1}^{n} T^r(\alpha_r,\alpha_{r-1}). \tag{103}$$

The diagonal del-operator MEs are the key to obtaining the off-diagonal MEs which are obtained via the use of the commutation relation

$$\Delta^m_{m+p} = [E^m_{m+p}, \Delta^{m+p}_{m+p}]. \tag{104}$$

The nonzero MEs of the off-diagonal operator $\Delta(n)^m_{m+p}$ are given by

$$\left\langle \begin{matrix} p_n \\ p_{n-1}+\epsilon_{\alpha_{n-1}} \\ \vdots \\ p_{m+p}+\epsilon_{\alpha_{m+p}} \\ p_{m+p-1}+\delta_{i_{m+p-1}} \\ \vdots \\ p_m+\delta_{i_m} \\ P_{m-1} \end{matrix} \right| \Delta(n)^m_{m+p} \left| \begin{matrix} p_n \\ p_{n-1} \\ \vdots \\ p_{m+p} \\ p_{m+p-1} \\ \vdots \\ p_m \\ P_{m-1} \end{matrix} \right\rangle = \Delta^* \begin{pmatrix} n-1 & & m+p-1 \\ & \cdots & \\ \alpha_{n-1} & & \alpha_{m+p-1} \end{pmatrix} \cdot V(\alpha_{m+p}, i_{m+p-1}) \cdot N \begin{pmatrix} m+p-1 & & m \\ & \cdots & \\ i_{m+p-1} & & i_m \end{pmatrix}. \tag{105}$$

In the above, Δ^* is as defined in Eq. (103) and

$$N \begin{pmatrix} m+p-1 & & m \\ & \cdots & \\ i_{m+p-1} & & i_m \end{pmatrix}$$

is the ME of the $U(n)$ generator E^m_{m+p} [see Eq. (75)].

The terms $V(\alpha_{m+p}, i_{m+p-1})$ are referred to as adjoint-vector coupling coefficients (AVCCs) and the lexically permitted possibilities are

$$\begin{aligned} V(-,2) &= (-1)^{\Delta a_{m+p}} \left(\frac{b_{m+p}+2\Delta bm+p-1}{b_{m+p-1}} \right)^{1/2} \cdot [(b_{m+p}+1)(b_{m+p}-1)]^{1/2}, \\ V(0,i_{m+p}) &= \tfrac{1}{2}\cdot(-1)^{i_{m+p-1}}(b_{m+p}+2\Delta b_{m+p}+2i_{m+p-1}-2), \\ V(+,1) &= (-1)^{\bar{\Delta} c_{m+p}} \left(\frac{3+b_{m+p}+2\Delta b_{m+p}}{b_{m+p}+3} \right)^{1/2} \cdot [(b_{m+p}+1)(b_{m+p}+3)]^{1/2}. \end{aligned} \tag{106}$$

In Eq. (106) we use the notation $\bar{\Delta} c_m = 1-\Delta c_m = 1-(c_m-c_{m-1})$.

The MEs of $\Delta(n+1)^{m+p}_m$ may be obtained by using the hermiticity requirement. They are stated explicitly by Gould and Paldus in Ref. 2.

As stated previously, rather than use the above shift component formalism for the multishell case, we will use the simpler approach based on coupling coefficients and the Racah factorization lemma. As an illustration of this method we will rederive the del operator ME of Eq. (105) using the ACC approach.

First, we show that zero-shift adjoint coupling coefficients may be defined via the del operator. That this is possible, follows from the unique vanishings property, Eq. (89), of this operator.

V. ZERO-SHIFT ADJOINT COUPLING COEFFICIENTS

We want to define zero-shift adjoint coupling coefficients (ACCs), that is those of the form

$$\left\langle \begin{matrix} p \\ P' \end{matrix} \middle| e_i \otimes \bar{e}_j ; \begin{matrix} p \\ P \end{matrix} \right\rangle, \tag{107}$$

where $\left|\begin{smallmatrix} p \\ P \end{smallmatrix}\right\rangle$ denotes a $U(n)$ GP basis state and $e_i \otimes \bar{e}_j \in V \otimes V^*$ is a GT basis state for the adjoint representation. The adjoint irrep $V \otimes V^*$, which has a highest weight of the form $\boldsymbol{\lambda}=(1,\dot{0},-1)$, will be denoted by $V(\boldsymbol{\lambda})=V(1,\dot{0},-1)$.

Now by a generalization of the discussion in Sec. III, the del operator Δ_{ij}, an adjoint tensor operator, is equivalent to an intertwining operator, Δ say, such that

$$\Delta : V(1,\dot{0},-1) \otimes \mathscr{H} \rightarrow \mathscr{H}, \tag{108}$$

where $\mathscr{H}$ is the Hilbert space of states

$$\mathscr{H} = \bigoplus_{\substack{p=(a,b,c) \\ a+b+c=n}} V(p). \tag{109}$$

The LHS of Eq. (108) contains subspaces of the form $V(1,\dot{0},-1) \otimes \mathrm{V(p)}$ which admit the Clebsch–Gordon decomposition (with only the lexically permitted subspaces shown)

$$V(1,\dot{0},-1) \otimes V(p) = V(p+\boldsymbol{\epsilon}) \oplus 2V(p) \oplus V(p-\boldsymbol{\epsilon}). \tag{110}$$

From Eq. (110) it is clear that there are multiple copies of the zero-shifted subspace.

From the decomposition of Eq. (110), we see that unique nonzero shift ACCs may be defined. But this is not true for the zero-shift case as here there is a multiplicity label. However, it is known that the RHS of Eq. (108), the Hilbert space $\mathscr{H}$, is multiplicity free and this fact makes the definition of zero-shift ACCs possible. We rewrite Eq. (108) to make this idea explicit

$$\Delta : V(1,\dot{0},-1) \otimes V(p) \rightarrow V(p+\boldsymbol{\epsilon}) \oplus V(p) \oplus V(p-\boldsymbol{\epsilon}) \subseteq \mathscr{H}. \tag{111}$$

In fact, the MEs of any zero-shift ATO may be used to define generalized coupling coefficients for the zero shift case. However, coupling coefficients so defined will not necessarily satisfy the Racah factorization lemma. As we will now show, the unique properties of the del operator make it the correct choice for the many electron problem.

From Gould and Battle[2] we know that the uniqueness of the vanishings property, Eq. (89), implies that the zero-shift component $\Delta\binom{n-1}{0}_{ij}$ of $\Delta(n)_{ij}$ is proportional to $\Delta(n-1)_{ij}$,

$$\Delta\begin{pmatrix} n-1 \\ 0 \end{pmatrix}_{ij} = (\alpha_{n-1}+1) \cdot \Delta(n-1)_{ij}, \quad 1 \leqslant i,j \leqslant n-1. \tag{112}$$

This fact implies that a unique factorization property for the del-operator MEs exists, namely,

$$\left\langle \begin{matrix} p_n \\ P'_{n-1} \end{matrix} \middle| \Delta(n)_{ij} \middle| \begin{matrix} p_n \\ P_{n-1} \end{matrix} \right\rangle = (\alpha_{n-1}+1) \cdot \left\langle \begin{matrix} p_{n-1} \\ P'_{n-2} \end{matrix} \middle| \Delta(n-1)_{ij} \middle| \begin{matrix} p_{n-1} \\ P_{n-2} \end{matrix} \right\rangle, \quad 1 \leqslant i,j \leqslant n-1. \tag{113}$$

This factorization may be continued provided the indices i and j are less than or equal to the index of the leading Paldus label

$$\left\langle \begin{matrix} p_n \\ p_{n-1} \\ p_{n-2} \\ P'_{n-3} \end{matrix} \middle| \Delta(n)_{ij} \middle| \begin{matrix} p_n \\ p_{n-1} \\ p_{n-2} \\ P_{n-3} \end{matrix} \right\rangle = (\alpha_{n-1}+1)(\alpha_{n-2}+1) \left\langle \begin{matrix} p_{n-2} \\ P'_{n-3} \end{matrix} \middle| \Delta(n-2)_{ij} \middle| \begin{matrix} p_{n-2} \\ P_{n-3} \end{matrix} \right\rangle, \quad 1 \leqslant i,j \leqslant n-2. \tag{114}$$

We now *define* the normalized del operator to be the zero-shift adjoint coupling coefficient, that is,

$$\left\langle \begin{matrix} p_n \\ P'_{n-1} \end{matrix} \middle| e_i \otimes \bar{e}_j ; \begin{matrix} p_n \\ P_{n-1} \end{matrix} \right\rangle \equiv \frac{1}{S(n)} \left\langle \begin{matrix} p_n \\ P'_{n-1} \end{matrix} \middle| \Delta(n)_{ij} \middle| \begin{matrix} p_n \\ P_{n-1} \end{matrix} \right\rangle, \quad 1 \leqslant i,j \leqslant n. \tag{115}$$

In the above $S(n)=-2S_n(S_n+1)$, where $S_n=b_n/2$ is the total spin. Note that $\mathrm{tr}[\Delta(n)]=S(n)$. This choice of normalization is required to ensure that (when $S \neq 0$)

$$\sum_{i,j,P''} \left\langle \begin{matrix} p \\ P' \end{matrix} \middle| e_i \otimes \bar{e}_j ; \begin{matrix} p \\ P'' \end{matrix} \right\rangle \left\langle \begin{matrix} p \\ P'' \end{matrix} \middle| e_i \otimes \bar{e}_j ; \begin{matrix} p \\ P \end{matrix} \right\rangle = \delta_{PP'}. \tag{116}$$

Now, if we use the zero-shift ACC definition of Eq. (115) in the factorization of Eq. (114) we get

$$S(n) \cdot \left\langle \begin{matrix} p_n \\ P'_{n-1} \end{matrix} \middle| e_i \otimes \bar{e}_j ; \begin{matrix} p_n \\ P_{n-1} \end{matrix} \right\rangle = (\alpha_{n-1}+1)(\alpha_{n-2}+1) \cdot S(n-2) \times \left\langle \begin{matrix} p_{n-2} \\ P'_{n-3} \end{matrix} \middle| e_i \otimes \bar{e}_j ; \begin{matrix} p_{n-2} \\ P_{n-3} \end{matrix} \right\rangle. \tag{117}$$

Rearranging and gathering terms we find that

$$\left\langle \begin{matrix} p_n \\ P'_{n-1} \end{matrix} \middle| e_i \otimes \bar{e}_j ; \begin{matrix} p_n \\ P_{n-1} \end{matrix} \right\rangle = \left[(\alpha_{n-1}+1) \frac{S(n-1)}{S(n)} \right] \cdot \left[(\alpha_{n-2}+1) \frac{S(n-2)}{S(n-1)} \right] \times \left\langle \begin{matrix} p_{n-2} \\ P'_{n-2} \end{matrix} \middle| e_i \otimes \bar{e}_j ; \begin{matrix} p_{n-2} \\ P_{n-2} \end{matrix} \right\rangle$$

or

$$\left\langle \begin{matrix} p_n \\ P'_{n-1} \end{matrix} \middle| e_i \otimes \bar{e}_j ; \begin{matrix} p_n \\ P_{n-1} \end{matrix} \right\rangle = \left\langle \begin{matrix} p_n \\ p_{n-1} \end{matrix} \middle\| \begin{matrix} p_n \\ p_{n-1} \end{matrix} \right\rangle \left\langle \begin{matrix} p_{n-1} \\ p_{n-2} \end{matrix} \middle\| \begin{matrix} p_{n-1} \\ p_{n-2} \end{matrix} \right\rangle \left\langle \begin{matrix} p_{n-2} \\ P'_{n-3} \end{matrix} \middle| e_i \otimes \bar{e}_j ; \begin{matrix} p_{n-2} \\ P_{n-3} \end{matrix} \right\rangle. \tag{118}$$

Coefficients of the form

$$\left\langle \begin{matrix} p_r \\ p_{r-1} \end{matrix} \middle\| \begin{matrix} p_r \\ p_{r-1} \end{matrix} \right\rangle$$

are examples of zero-shift adjoint reduced Wigner coefficients (RWCs). They depend only on the Paldus labels p_r and p_{r-1} of the subgroups $U(r)$ and $U(r-1)$. Equations (118) constitutes the Racah factorization lemma for the zero-shift case.

We now need to demonstrate the factorization property for the case where one of the shifts (at level r or $r-1$) is zero and the shift at the other level is nonzero. The factorization property for those of the form where both levels are shifted (viz. $p'_r = p_r \pm \epsilon$ and $p'_{r-1} = p_{r-1} \pm \epsilon$) follows immediately from the uniqueness of the nonzero adjoint coupling coefficients.

For convenience we work in an irrep of $U(n+1)$.

Case 1

When $p'_{n+1} = p_{n+1}$ and $p'_n = p_n \pm \epsilon$.

From the definition of the zero-shift adjoint coupling coefficients we have

$$\left\langle \begin{matrix} p_{n+1} \\ p_n \pm \epsilon \\ P'_{n-1} \end{matrix} \middle| \hat{\Delta}_{ij} \middle| \begin{matrix} p_{n+1} \\ p_n \\ P_{n-1} \end{matrix} \right\rangle = S(n+1) \cdot \left\langle \begin{matrix} p_{n+1} \\ p_n \pm \epsilon \\ P'_{n-1} \end{matrix} \middle| e_i \otimes \bar{e}_j ; \begin{matrix} p_{n+1} \\ p_n \\ P_{n-1} \end{matrix} \right\rangle . \tag{119}$$

Now, from Gould and Paldus,[1] the application of the $U(n)$ WE theorem to the lhs of Eq. (119) yields

$$\left\langle \begin{matrix} p_{n+1} \\ p_n \pm \epsilon \\ P'_{n-1} \end{matrix} \middle| \hat{\Delta}_{ij} \middle| \begin{matrix} p_{n+1} \\ p_n \\ P_{n-1} \end{matrix} \right\rangle = \langle p_n \pm \epsilon \| \hat{\Delta} \| p_n \rangle \cdot \left\langle \begin{matrix} p_n \pm \epsilon \\ P'_{n-1} \end{matrix} \middle| e_i \otimes \bar{e}_j ; \begin{matrix} p_n \\ P_{n-1} \end{matrix} \right\rangle . \tag{120}$$

In both of the above equations we have $1 \leqslant i,j \leqslant n$. Equating Eqs. (119) and (120) and collecting terms we have

$$\begin{aligned} \left\langle \begin{matrix} p_{n+1} \\ p_n \pm \epsilon \\ P'_{n-1} \end{matrix} \middle| e_i \otimes \bar{e}_j ; \begin{matrix} p_{n+1} \\ p_n \\ P_{n-1} \end{matrix} \right\rangle &= \frac{\langle p_n \pm \epsilon \| \hat{\Delta} \| p_n \rangle}{S(n+1)} \cdot \left\langle \begin{matrix} p_n \pm \epsilon \\ P'_{n-1} \end{matrix} \middle| e_i \otimes \bar{e}_j ; \begin{matrix} p_n \\ P_{n-1} \end{matrix} \right\rangle \\ &= \left\langle \begin{matrix} p_{n+1} \\ p_n \pm \epsilon \end{matrix} \middle\| \begin{matrix} p_{n+1} \\ p_n \end{matrix} \right\rangle \cdot \left\langle \begin{matrix} p_n \pm \epsilon \\ P'_{n-1} \end{matrix} \middle| e_i \otimes \bar{e}_j ; \begin{matrix} p_n \\ P_{n-1} \end{matrix} \right\rangle . \end{aligned} \tag{121}$$

Case 2

When $p'_n = p_n$ and $p'_{n+1} = p_{n+1} \pm \epsilon$.

The factorization property of the del operator enables us to say that

$$\left\langle \begin{matrix} p_{n+1} \pm \epsilon \\ p_n \\ P'_{n-1} \end{matrix} \middle| \hat{\Delta}_{ij} \middle| \begin{matrix} p_{n+1} \\ p_n \\ P_{n-1} \end{matrix} \right\rangle = \gamma_{n+1} \cdot \left\langle \begin{matrix} p_n \\ P'_{n-1} \end{matrix} \middle| \Delta_{ij} \middle| \begin{matrix} p_n \\ P_{n-1} \end{matrix} \right\rangle , \tag{122}$$

where the proportionality constant γ_{n+1} is a $U(n)$ invariant. Now, by definition

$$\left\langle \begin{matrix} p_n \\ P'_{n-1} \end{matrix} \middle| \Delta_{ij} \middle| \begin{matrix} p_n \\ P_{n-1} \end{matrix} \right\rangle = S(n) \cdot \left\langle \begin{matrix} p_n \\ P'_{n-1} \end{matrix} \middle| e_i \otimes \bar{e}_j ; \begin{matrix} p_n \\ P_{n-1} \end{matrix} \right\rangle . \tag{123}$$

And from the $U(n+1)$ WE theorem

$$\begin{aligned} \left\langle \begin{matrix} p_{n+1} \pm \epsilon \\ p_n \\ P'_{n-1} \end{matrix} \middle| \hat{\Delta}_{ij} \middle| \begin{matrix} p_{n+1} \\ p_n \\ P_{n-1} \end{matrix} \right\rangle \\ = \langle p_{n+1} \pm \epsilon \| \hat{\Delta} \| p_{n+1} \rangle \cdot \left\langle \begin{matrix} p_{n+1} \pm \epsilon \\ p_n \\ P'_{n-1} \end{matrix} \middle| e_i \otimes \bar{e}_j ; \begin{matrix} p_{n+1} \\ p_n \\ P_{n-1} \end{matrix} \right\rangle . \end{aligned} \tag{124}$$

Equations (122), (123), and (124) yield

$$\begin{aligned} \left\langle \begin{matrix} p_{n+1} \pm \epsilon \\ p_n \\ P'_{n-1} \end{matrix} \middle| e_i \otimes \bar{e}_j ; \begin{matrix} p_{n+1} \\ p_n \\ P_{n-1} \end{matrix} \right\rangle \\ = \frac{\gamma \cdot S(n)}{\langle p_{n+1} \pm \epsilon \| \hat{\Delta} \| p_{n+1} \rangle} \cdot \left\langle \begin{matrix} p_n \\ P'_{n-1} \end{matrix} \middle| e_i \otimes \bar{e}_j ; \begin{matrix} p_n \\ P_{n-1} \end{matrix} \right\rangle \\ = \left\langle \begin{matrix} p_{n+1} \pm \epsilon \\ p_n \end{matrix} \middle\| \begin{matrix} p_{n+1} \\ p_n \end{matrix} \right\rangle \left\langle \begin{matrix} p_n \\ P'_{n-1} \end{matrix} \middle| e_i \otimes \bar{e}_j ; \begin{matrix} p_n \\ P_{n-1} \end{matrix} \right\rangle . \end{aligned} \tag{125}$$

To summarize, we have the basic factorization property

$$\begin{aligned} \left\langle \begin{matrix} p_r + \epsilon_{\alpha_r} \\ p_{r-1} + \epsilon_{\alpha_{r-1}} \\ P'_{r-2} \end{matrix} \middle| e_i \otimes \bar{e}_j ; \begin{matrix} p_r \\ p_{r-1} \\ P_{r-2} \end{matrix} \right\rangle \\ = \left\langle \begin{matrix} p_r + \epsilon_{\alpha_r} \\ p_{r-1} + \epsilon_{\alpha_{r-1}} \end{matrix} \middle\| \begin{matrix} p_r \\ p_{r-1} \end{matrix} \right\rangle \cdot \left\langle \begin{matrix} p_{r-1} + \epsilon_{\alpha_{r-1}} \\ P'_{r-2} \end{matrix} \middle| e_i \otimes \bar{e}_j ; \begin{matrix} p_{r-1} \\ P_{r-2} \end{matrix} \right\rangle , \end{aligned} \tag{126}$$

where $\epsilon_{\alpha_r} = +\epsilon, 0, -\epsilon$ and $1 \leqslant i, j \leqslant n$. Equation (126) is the Racah factorization lemma for both zero and nonzero shift ACCs.

With this definition of the zero-shift ACCs, Eq. (115), we may now adopt a unified approach to the evaluation of the MEs of the del operator. Since the $U(m)$ del operator $\Delta(m)_{ij}$ affects a zero shift at the top Paldus label p_m, we may write

$$\left\langle \begin{matrix} p_m \\ P'_{m-1} \end{matrix} \middle| \Delta(m)_{ij} \middle| \begin{matrix} p_m \\ P_m \end{matrix} \right\rangle = S(m) \left\langle \begin{matrix} p_m \\ P'_{m-1} \end{matrix} \middle| e_i \otimes \bar{e}_j ; \begin{matrix} p_m \\ P_{m-1} \end{matrix} \right\rangle \tag{127}$$

for all types of del operator and then apply the Racah factorization lemma to express the del-operator MEs entirely in terms of reduced Wigner coefficients and Wigner coefficients. Furthermore, as we will show in the next section, this factorization is directly equivalent to the segmentation level formulas obtained by Gould and Battle.[2]

VI. THE DEL-OPERATOR MATRIX ELEMENTS

In this section we derive the MEs of the del operators of the form $\Delta(n)_{m,m+p}$ using the Racah factorization lemma. Our development parallels that of Gould and Battle[2] to allow a direct comparison with the shift operator method. Thus our presentation has three key steps:

(i) Demonstrate that the Racah factorization yields the basic segmentation level formulae of Eqs. (101).
(ii) Extend the reduction to obtain the MEs of the diagonal del-operators $\Delta(n)_{mm}$, Eq. (102).
(iii) Evaluate the general off-diagonal del-operator MEs of the form $\Delta(n)_{m,m+p}$, Eq. (105).

A. The basic segmentation level formulas

For the work in this section we need the adjoint reduced Wigner coefficients. These are of the form

$$\left\langle \begin{matrix} p_n+\epsilon_{\alpha_n} \\ p_{n-1}+\epsilon_{\alpha_{n-1}} \end{matrix} \middle\| \begin{matrix} p_n \\ p_{n-1} \end{matrix} \right\rangle. \tag{128}$$

We note that the shifts $(\epsilon_{\alpha_n},\epsilon_{\alpha_{n-1}})=(+,-)$ or $(-,+)$ are nonlexical and thus do not occur. Using the methods of Gould *et al.*,[4–9] these coefficients are derived in the second and third papers of this series. The lexically permitted adjoint RWCs are tabulated in Appendix D. Our purpose in this section is to take these coefficients as given and use them to rederive the del-operator matrix elements via the ACC approach.

To parallel the work of Gould and Battle,[2] we first work in an irrep of $U(n+1)$. Now, recalling that the del operator is an ATO and effects a zero shift on the $U(n+1)$ label p_{n+1}, the first step in the calculation of the ME is always to equate it to the corresponding adjoint Wigner coefficient. The application of the Racah factorization lemma, and the simplification of the resulting expression by use of the corresponding level shifts, yields the required segment level formula.

We will illustrate with some sample calculations.

(i)

$$\left\langle \begin{matrix} p_{n+1} \\ p_n \\ p_{n-1} \\ P_{n-2} \end{matrix} \middle| \Delta\begin{pmatrix} n & n-1 \\ 0 & 0 \end{pmatrix}_{n-1,n-1} \middle| \begin{matrix} p_{n+1} \\ p_n \\ p_{n-1} \\ P_{n-2} \end{matrix} \right\rangle$$

$$=S(n+1)\cdot\left\langle \begin{matrix} p_{n+1} \\ p_n \\ p_{n-1} \\ P_{n-2} \end{matrix} \middle| e_{n-1}\otimes\bar{e}_{n-1}; \begin{matrix} p_{n+1} \\ p_n \\ p_{n-1} \\ P_{n-2} \end{matrix} \right\rangle = S(n+1)\left\langle \begin{matrix} p_{n+1} \\ p_n \end{matrix} \middle\| \begin{matrix} p_{n+1} \\ p_n \end{matrix} \right\rangle \left\langle \begin{matrix} p_n \\ p_{n-1} \end{matrix} \middle\| \begin{matrix} p_n \\ P_{n-1} \end{matrix} \right\rangle \left\langle \begin{matrix} p_{n-1} \\ P_{n-2} \end{matrix} \middle| e_{n-1}\otimes\bar{e}_{n-1}; \begin{matrix} p_{n-1} \\ P_{n-2} \end{matrix} \right\rangle$$

$$=S(n+1)\cdot\left[\frac{S(n)}{S(n+1)}\cdot(\alpha_n+1)\right]\cdot\left[\frac{S(n-1)}{S(n)}\cdot(\alpha_{n-1}+1)\right]\cdot\frac{\Delta(n-1)_{n-1,n-1}}{S(n-1)}$$

$$=(\alpha_n+1)\cdot(\alpha_{n-1}+1)\cdot\Delta(n-1)_{n-1,n-1}=T^{n+1}(0)\cdot T^n(0,0)\cdot T_{n-1}(0). \tag{129}$$

(ii)

$$\left\langle \begin{matrix} p_{n+1} \\ p_n+\epsilon_- \\ p_{n-1} \\ P_{n-2} \end{matrix} \middle| \Delta\begin{pmatrix} n & n-1 \\ - & 0 \end{pmatrix}_{n-1,n-1} \middle| \begin{matrix} p_{n+1} \\ p_n \\ p_{n-1} \\ P_{n-2} \end{matrix} \right\rangle$$

$$=S(n+1)\cdot\left\langle \begin{matrix} p_{n+1} \\ p_n \\ p_{n-1} \\ P_{n-2} \end{matrix} \middle| e_{n-1}\otimes\bar{e}_{n-1}; \begin{matrix} p_{n+1} \\ p_n \\ p_{n-1} \\ P_{n-2} \end{matrix} \right\rangle = S(n+1)\cdot\left\langle \begin{matrix} p_{n+1} \\ p_n+\epsilon_- \end{matrix} \middle\| \begin{matrix} p_{n+1} \\ p_n \end{matrix} \right\rangle\cdot\left\langle \begin{matrix} p_n+\epsilon_- \\ p_{n-1} \end{matrix} \middle\| \begin{matrix} p_n \\ p_{n-1} \end{matrix} \right\rangle\cdot\left\langle \begin{matrix} p_{n-1} \\ P_{n-2} \end{matrix} \middle| e_{n-1}\otimes\bar{e}_{n-1}; \begin{matrix} p_{n-1} \\ P_{n-2} \end{matrix} \right\rangle$$

$$=S(n+1)\cdot\left[\frac{1}{S(n+1)}\cdot\left\{\frac{(a_n+1)(b_n+1)(c_n+1)}{b_n}\right\}^{1/2}\right]\left[(-1)\left\{\frac{b_{n-1}}{(a_{n-1}+1)(b_{n-1}+1)(c_{n-1}+1)}\right\}^{1/2}\right]\cdot\frac{\Delta(n-1)_{n-1,n-1}}{S(n-1)}.$$

Using the shifts $p_{n+1}=p_n+(1,-1,1)$ and $p_n=p_{n-1}+(0,1,0)$ this becomes

$$=\frac{1}{1+b_{n+1}}\cdot 2[(1+b_n)(1+b_n-2)]^{-1/2}\cdot\Delta(n-1)_{n-1,n-1}=T^{n+1}(-)\cdot T^n(-,0)\cdot T_{n-1}(0). \tag{130}$$

(iii)

$$\left\langle \begin{matrix} p_{n+1} \\ p_n \\ p_{n-1}+\epsilon_+ \\ P_{n-2} \end{matrix} \right| \Delta\begin{pmatrix} n & n-1 \\ 0 & + \end{pmatrix}_{n-1,n-1} \left| \begin{matrix} p_{n+1} \\ p_n \\ p_{n-1} \\ P_{n-2} \end{matrix} \right\rangle$$

$$=S(n+1)\cdot \left\langle \begin{matrix} p_{n+1} \\ p_n \\ p_{n-1} \\ P_{n-2} \end{matrix} \right| e_{n-1}\otimes\bar{e}_{n-1}; \begin{matrix} p_{n+1} \\ p_n \\ p_{n-1} \\ P_{n-2} \end{matrix} \right\rangle = S(n+1)\cdot \left\langle \begin{matrix} p_{n+1} \\ p_n \end{matrix} \right\| \left| \begin{matrix} p_{n+1} \\ p_n \end{matrix} \right\rangle \cdot \left\langle \begin{matrix} p_n \\ p_{n-1}+\epsilon_+ \end{matrix} \right\| \left| \begin{matrix} p_n \\ p_{n-1} \end{matrix} \right\rangle \cdot \left\langle \begin{matrix} p_{n-1} \\ P_{n-2} \end{matrix} \right| e_{n-1}\otimes\bar{e}_{n-1}; \begin{matrix} p_{n-1} \\ P_{n-2} \end{matrix} \right\rangle . \tag{131}$$

Now, the full adjoint WC on the RHS of Eq. (131) is equal (up to a phase) to the corresponding RWC, see Gould and Paldus.[9] That is,

$$\left\langle \begin{matrix} p_{n-1}+\epsilon_+ \\ P_{n-2} \end{matrix} \right| e_{n-1}\otimes\bar{e}_{n-1}; \begin{matrix} p_{n-1} \\ P_{n-2} \end{matrix} \right\rangle = (-1) \left\langle \begin{matrix} p_{n-1}+\epsilon_+ \\ p_{n-2} \end{matrix} \right\| \left| \begin{matrix} p_{n-1} \\ p_{n-2} \end{matrix} \right\rangle . \tag{132}$$

In Eq. (132) p_{n-2} is the leading Paldus label of the Paldus array P_{n-2}. The phase change is required to ensure that the maximal WCs have a positive real phase.

After substitution we get for Eq. (131)

$$\left\langle \begin{matrix} p_{n+1} \\ p_n \\ p_{n-1}+\epsilon_+ \\ P_{n-2} \end{matrix} \right| \Delta\begin{pmatrix} n & n-1 \\ 0 & + \end{pmatrix}_{n-1,n-1} \left| \begin{matrix} p_{n+1} \\ p_n \\ p_{n-1} \\ P_{n-2} \end{matrix} \right\rangle$$

$$=S(n+1)\cdot\left[\frac{S(n)}{S(n+1)}\cdot(\alpha_n+1)\right]\left[\frac{1}{S(n)}\cdot\left\{\frac{(\overline{n-1}+2-c_{n-1})(b_{n-1}+1)(\overline{n-1}+2-a_{n-1})}{(2+b_{n-1})}\right\}^{1/2}\right]$$

$$\times\left[\left\{\frac{(b_{n-2}+2)}{(b_{n-2}+1)(\overline{n-2}+2-c_{n-2})(\overline{n-2}+2-a_{n-2})}\right\}^{1/2}\right].$$

Application of the shifts $p_n=p_{n-1}+(0,1,0)$ and $p_{n-1}=p_{n-2}+(1,-1,1)$ and rearrangement reduces this to

$$=(\alpha_n+1)\cdot\frac{1}{1+b_n}\cdot[(1+b_{n-1})\cdot(1+b_{n-1}+2)]^{1/2}=T^{n+1}(0)\cdot T^n(0,+)\cdot T_{n-1}(+). \tag{133}$$

The adjoint RWCs

$$\left\langle \begin{matrix} p_n+\epsilon_+ \\ p_{n-1}+\epsilon_+ \end{matrix} \right\| \left| \begin{matrix} p_n \\ p_{n-1} \end{matrix} \right\rangle \quad \text{and} \quad \left\langle \begin{matrix} p_n+\epsilon_- \\ p_{n-1}+\epsilon_- \end{matrix} \right\| \left| \begin{matrix} p_n \\ p_{n-1} \end{matrix} \right\rangle$$

are shift dependent and the corresponding ME formulas must be obtained on a case by case basis. For the $(+,+)$ shifts we have (iv)

$$\left\langle \begin{matrix} p_{n+1} \\ p_n+\epsilon_+ \\ p_{n-1}+\epsilon_+ \\ P_{n-2} \end{matrix} \right| \Delta\begin{pmatrix} n & n-1 \\ + & + \end{pmatrix}_{n-1,n-1} \left| \begin{matrix} p_{n+1} \\ p_n \\ p_{n-1} \\ P_{n-2} \end{matrix} \right\rangle$$

$$=S(n+1)\cdot \left\langle \begin{matrix} p_{n+1} \\ p_n+\epsilon_+ \\ p_{n-1}+\epsilon_+ \\ P_{n-2} \end{matrix} \right| e_{n-1}\otimes\bar{e}_{n-1}; \begin{matrix} p_{n+1} \\ p_n \\ p_{n-1} \\ P_{n-2} \end{matrix} \right\rangle$$

$$=S(n+1)\cdot \left\langle \begin{matrix} p_{n+1} \\ p_n+\epsilon_+ \end{matrix} \right\| \left| \begin{matrix} p_{n+1} \\ p_n \end{matrix} \right\rangle \cdot \left\langle \begin{matrix} p_n+\epsilon_+ \\ p_{n-1}+\epsilon_+ \end{matrix} \right\| \left| \begin{matrix} p_n \\ p_{n-1} \end{matrix} \right\rangle \cdot \left\langle \begin{matrix} p_n+\epsilon_+ \\ P_{n-2} \end{matrix} \right| e_{n-1}\otimes\bar{e}_{n-1}; \begin{matrix} p_{n-1} \\ P_{n-2} \end{matrix} \right\rangle$$

$$=S(n+1)\cdot\left[\frac{1}{S(n+1)}\cdot\left\{\frac{(n+2-c_n)(b_n+1)(n+2-a_n)}{(2+b_n)}\right\}^{1/2}\right]\cdot\left\langle \begin{matrix} p_n+\epsilon_+ \\ p_{n-1}+\epsilon_+\end{matrix}\middle\|\begin{matrix} p_n \\ p_{n-1}\end{matrix}\right\rangle$$

$$\times\left[\frac{(b_{n-2}+2)}{(b_{n-2}+1)(\overline{n-2}+2-c_{n-2})(\overline{n-2}+2-a_{n-1})}\right]. \tag{134}$$

The four subcases of the RWC $\langle{}^{p_n+\epsilon_+}_{p_{n-1}+\epsilon_+}\|{}^{p_n}_{p_{n-1}}\rangle$ are listed in Appendix D. Substitution, on a case by case basis, together with the use of the level shifts $p_{n+1}=p_n+(0,1,0)$ and $p_{n-1}=p_{n-2}+(1,-1,1)$ gives

$$\left\langle \begin{matrix} p_{n+1} \\ p_n+\epsilon_+ \\ p_{n-1}+\epsilon_+ \\ P_{n-2}\end{matrix}\middle| \Delta\begin{pmatrix} n & n-1 \\ + & + \end{pmatrix}_{n-1,n-1}\middle| \begin{matrix} p_{n+1} \\ p_n \\ p_{n-1} \\ P_{n-2}\end{matrix}\right\rangle$$

$$=\begin{cases} \dfrac{1}{1+b_{n+1}}\cdot(+1)\cdot[(1+b_{n-1})(1+b_{n-1}+2)]^{1/2}, & \Delta b_n=0, \\ \dfrac{1}{1+b_{n+1}}\cdot(-1)\cdot\left(\dfrac{b_n+3}{b_n+1}\right)^{1/2}[(1+b_{n-1})(1+b_{n-1}+2)]^{1/2}, & \Delta b_n=1, \\ \dfrac{1}{1+b_{n+1}}\cdot(-1)\cdot\left(\dfrac{b_n+1}{b_n+3}\right)^{1/2}[(1+b_{n-1})(1+b_{n-1}+2)]^{1/2}, & \Delta b_n=-1. \end{cases} \tag{135}$$

We will not reproduce the full working, but a similar case by case evaluation of the corresponding segment level formula of Gould and Battle[2] shows that the two formulas are identically equal

$$\left\langle \begin{matrix} p_{n+1} \\ p_n+\epsilon_+ \\ p_{n-1}+\epsilon_+ \\ P_{n-2}\end{matrix}\middle| \Delta\begin{pmatrix} n & n-1 \\ + & + \end{pmatrix}_{n-1,n-1}\middle| \begin{matrix} p_{n+1} \\ p_n \\ p_{n-1} \\ P_{n-2}\end{matrix}\right\rangle = T^{n+1}(+)\cdot T^n(+,+)\cdot T_{n-1}(+). \tag{136}$$

Similar results hold for all of the remaining MEs and the segmentation level formulas of Eqs. (101) are reproduced exactly in all cases by the use of the Racah factorization lemma.

B. The diagonal matrix elements

We want to determine the MEs of the diagonal del operators of the form $\Delta(n+1)_{mm}$. To do so we may extend the above procedure which is essentially a recursive reduction. As before

$$\left\langle \begin{matrix} p_{n+1} \\ p_n+\epsilon_{\alpha_n} \\ \vdots \\ p_m+\epsilon_{\alpha_m} \\ P_{m-1}\end{matrix}\middle| \Delta(n+1)_{mm} \middle| \begin{matrix} p_{n+1} \\ p_n \\ \cdots \\ p_m \\ P_{m-1}\end{matrix}\right\rangle$$

$$=S(n+1)\cdot\left\langle \begin{matrix} p_{n+1} \\ p_n+\epsilon_{\alpha_n} \\ \vdots \\ p_m+\epsilon_{\alpha_m} \\ P_{m-1}\end{matrix}\middle| e_m\otimes\bar{e}_m; \begin{matrix} p_{n+1} \\ p_n \\ \cdots \\ p_m \\ P_{m-1}\end{matrix}\right\rangle. \tag{137}$$

Application of the Racah factorization lemma down to the mth level yields

$$=S(n+1)\cdot\left\langle \begin{matrix} p_{n+1} \\ p_n+\epsilon_{\alpha_n}\end{matrix}\middle\|\begin{matrix} p_{n+1} \\ p_n\end{matrix}\right\rangle$$

$$\times\prod_{r=m+1}^{n}\left\langle \begin{matrix} p_r+\epsilon_{\alpha_r} \\ p_{r-1}+\epsilon_{\alpha_{r-1}}\end{matrix}\middle|\times\middle|\begin{matrix} p_r \\ p_{r-1}\end{matrix}\right\rangle$$

$$\times\left\langle \begin{matrix} p_m+\epsilon_{\alpha_m} \\ P_{m-1}\end{matrix}\middle| e_m\otimes\bar{e}_m; \begin{matrix} p_m \\ P_{m-1}\end{matrix}\right\rangle. \tag{138}$$

At this stage, to simplify the working, we need a more explicit relationship between the individual adjoint RWCs and the corresponding segment level formulas. From an analysis of Eqs. (129) to (136) (and the other cases not shown) we may infer the general relationship

$$\left\langle \begin{matrix} p_r+\epsilon_{\alpha_r} \\ p_{r-1}+\epsilon_{\alpha_{r-1}}\end{matrix}\middle\|\begin{matrix} p_r \\ p_{r-1}\end{matrix}\right\rangle$$

$$=\frac{1}{P_{\alpha_r}(r)}\cdot T^r(\alpha_r,\alpha_{r-1})\cdot P_{\alpha_{r-1}}(r-1) \tag{139}$$

and

$$\left\langle \begin{matrix} p_m+\epsilon_{\alpha_m} \\ P_{m-1}\end{matrix}\middle| e_m\otimes\bar{e}_m; \begin{matrix} p_m \\ P_{m-1}\end{matrix}\right\rangle = \left\langle \begin{matrix} p_m+\epsilon_{\alpha_m} \\ p_{m-1}\end{matrix}\middle\|\begin{matrix} p_m \\ p_{m-1}\end{matrix}\right\rangle$$

$$= \frac{1}{P_{\alpha_m}(m)} \cdot T_m(\alpha_m). \tag{140}$$

The expressions $P_{\alpha_n}(n)$ are given by

$$P_+(n) = [(b_n+1)(b_n+2)(n+2-a_n)(n+2-c_n)]^{1/2},$$
$$P_-(n) = [b_n(a_n+1)(b_n+1)(c_n+1)]^{1/2}, \tag{141}$$
$$P_0(n) = S(n) = -2S_n(S_n+1).$$

Equations (139) to (141) express the equivalence of the factorization approach to the shift operator approach much more directly. Noting that $T^{n+1}(0) = T^{n+1}(0,0)$ and $T^{n+1}(\pm) = T^{n+1}(0,\pm)$, substitution of Eqs. (139) and (140) into Eq. (138) gives

$$\left\langle \begin{matrix} p_{n+1} \\ p_n + \epsilon_{\alpha_n} \\ \vdots \\ p_m + \epsilon_{\alpha_m} \\ P_{m-1} \end{matrix} \middle| \Delta(n+1)_{mm} \middle| \begin{matrix} p_{n+1} \\ p_n \\ \vdots \\ p_m \\ P_{m-1} \end{matrix} \right\rangle$$
$$= S(n+1) \cdot \prod_{r=m+1}^{n+1} \left\langle \begin{matrix} p_r + \epsilon_{\alpha_r} \\ p_{r-1} + \epsilon_{\alpha_{r-1}} \end{matrix} \middle\| \begin{matrix} p_r \\ p_{r-1} \end{matrix} \right\rangle$$
$$\times \left\langle \begin{matrix} p_m + \epsilon_{\alpha_m} \\ p_{m-1} \end{matrix} \middle\| \begin{matrix} p_m \\ p_{m-1} \end{matrix} \right\rangle$$
$$= \prod_{r=m+1}^{n+1} T^r(\alpha_r, \alpha_{r-1}) P_{\alpha_m}(m) \cdot \frac{1}{P_{\alpha_m}(m)} T_m(\alpha_m)$$
$$= T^{n+1}(\alpha_n) \cdot \prod_{r=m+1}^{n} T^r(\alpha_r, \alpha_{r-1}) \cdot T_m(\alpha_m)$$
$$= \Delta^* \begin{pmatrix} n & & m+1 \\ & \cdots & \\ \alpha_n & & \alpha_{m+1} \end{pmatrix} \cdot T_m(\alpha_m). \tag{142}$$

This defines Δ^* [as for Eq. (103)] by

$$\Delta^* \begin{pmatrix} n & & m+1 \\ & \cdots & \\ \alpha_n & & \alpha_{m+1} \end{pmatrix}$$
$$= T^{n+1}(\alpha_m) \cdot \prod_{r=m+1}^{n} T^r(\alpha_r, \alpha_{r-1}). \tag{143}$$

Equation (142) agrees identically with the formula of Gould and Battle.[2] The replacement $n+1 \mapsto n$ will give the ME of $\Delta(n)_{mm}$.

C. The matrix elements of $\Delta(n)_{m,m+p}$

The nonvanishing MEs of the operator $\Delta(n)_{m,m+p}$ are found to be,[2,5]

$$\left\langle \begin{matrix} p_n \\ p_{n-1} + \epsilon_{\alpha_{n-1}} \\ \vdots \\ p_{m+p} + \epsilon_{\alpha_{m+p}} \\ p_{m+p-1} + \delta_{i_{m+p-1}} \\ \vdots \\ p_m + \delta_{i_m} \\ P_{m-1} \end{matrix} \middle| \Delta(n)_{m,m+p} \middle| \begin{matrix} p_n \\ p_{n-1} \\ \vdots \\ p_{m+p} \\ p_{m+p-1} \\ \vdots \\ p_m \\ P_{m-1} \end{matrix} \right\rangle$$
$$= S(n) \cdot \left\langle \begin{matrix} p_n \\ p_{n-1} + \epsilon_{\alpha_{n-1}} \\ \vdots \\ p_{m+p} + \epsilon_{\alpha_{m+p}} \\ p_{m+p-1} + \delta_{i_{m+p-1}} \\ \vdots \\ p_m + \delta_{i_m} \\ P_{m-1} \end{matrix} \middle| e_m \otimes \bar{e}_{m+p}; \begin{matrix} p_n \\ p_{n-1} \\ \vdots \\ p_{m+p} \\ p_{m+p-1} \\ \vdots \\ p_m \\ P_{m-1} \end{matrix} \right\rangle \tag{144}$$

and after application of the Racah factorization lemma this becomes

$$= S(n) \cdot \left\langle \begin{matrix} p_n \\ p_{n-1} + \epsilon_{\alpha_{n-1}} \end{matrix} \middle\| \begin{matrix} p_n \\ p_{n-1} \end{matrix} \right\rangle \prod_{r=m+p}^{n-2} \left\langle \begin{matrix} p_{r+1} + \epsilon_{\alpha_{r+1}} \\ p_r + \epsilon_{\alpha_r} \end{matrix} \middle\| \begin{matrix} p_{r+1} \\ p_r \end{matrix} \right\rangle$$
$$\times \left\langle \begin{matrix} p_{m+p} + \epsilon_{\alpha_{m+p}} \\ p_{m+p-1} + \delta_{i_{m+p-1}} \end{matrix} \middle\| \begin{matrix} p_{m+p} \\ p_{m+p-1} \end{matrix} \right\rangle$$
$$\times \left\langle \begin{matrix} p_{m+p-1} + \delta_{i_{m+p-1}} \\ \vdots \\ p_m + \delta_{i_m} \\ P_{m-1} \end{matrix} \middle| e_m; \begin{matrix} p_{m+p-1} \\ \vdots \\ p_m \\ P_{m-1} \end{matrix} \right\rangle. \tag{145}$$

The coefficient

$$\left\langle \begin{matrix} p_{m+p} + \epsilon_{\alpha_{m+p}} \\ p_{m+p-1} + \delta_{i_{m+p-1}} \end{matrix} \middle\| \begin{matrix} p_{m+p} \\ p_{m+p-1} \end{matrix} \right\rangle \tag{146}$$

is referred to as an adjoint-vector RWC. The lexically permitted adjoint-vector RWCs are tabulated in Appendix E. They are obtained by the same methods as used for the adjoint RWCs.

The last term in Eq. (145) is a VCC which is given by

$$\left\langle \begin{matrix} p_{m+p-1} + \delta_{i_{m+p-1}} \\ \vdots \\ p_m + \delta_{i_m} \\ P_{m-1} \end{matrix} \middle| e_m; \begin{matrix} p_{m+p-1} \\ \vdots \\ p_m \\ P_{m-1} \end{matrix} \right\rangle$$
$$= V \begin{pmatrix} i_{m+p-1} & & i_m \\ & \cdots & \\ m_p - 1 & & m \end{pmatrix}$$
$$= [R^{i_{m+p-1}}_{m+p-1}]^{-1/2} \cdot N \begin{pmatrix} i_{m+p-1} & & i_m \\ & \cdots & \\ m+p-1 & & m \end{pmatrix}, \tag{147}$$

where in the last step we have applied the $U(n)$ WE theorem of Eq. (79).

From the previous section we have that

$$S(n)\cdot\left\langle \begin{matrix} p_n \\ p_{n-1}+\epsilon_{\alpha_n} \end{matrix} \middle\| \begin{matrix} p_n \\ p_{n-1} \end{matrix} \right\rangle \cdot \prod_{r=m+p}^{n-2} \left\langle \begin{matrix} p_{r+1}+\epsilon_{\alpha_{r+1}} \\ p_r+\epsilon_{\alpha_r} \end{matrix} \middle\| \begin{matrix} p_{r+1} \\ p_r \end{matrix} \right\rangle$$

$$=\Delta^*\begin{pmatrix} n-1 & & m+p-1 \\ & \cdots & \\ \alpha_{n-1} & & \alpha_{m+p-1} \end{pmatrix}\cdot P_{\alpha_{m+p}}(m+p). \quad (148)$$

Finally, the substitution of Eqs. (147) and (148) into Eq. (145) yields

$$\left\langle \begin{matrix} p_n \\ p_{n-1}+\epsilon_{\alpha_{n-1}} \\ \vdots \\ p_{m+p}+\epsilon_{\alpha_{m+p}} \\ p_{m+p-1}+\delta_{i_{m+p-1}} \\ \vdots \\ p_m+\delta_{i_m} \\ P_{m-1} \end{matrix} \middle| \Delta(n)_{m,m+p} \middle| \begin{matrix} p_n \\ p_{n-1} \\ \vdots \\ p_{m+p} \\ p_{m+p-1} \\ \vdots \\ p_m \\ P_{m-1} \end{matrix} \right\rangle$$

$$=\Delta^*\begin{pmatrix} n-1 & & m+p+1 \\ & \cdots & \\ \alpha_{n-1} & & \alpha_{m+p+1} \end{pmatrix}\cdot P_{\alpha_{m+p}}(m+p)$$

$$\times[R^{i_{m+p-1}}_{m+p-1}]^{-1/2}\left\langle \begin{matrix} p_{m+p}+\epsilon_{\alpha_{m+p}} \\ p_{m+p-1}+\delta_{i_{m+p-1}} \end{matrix} \middle\| \begin{matrix} p_{m+p} \\ p_{m+p-1} \end{matrix} \right\rangle$$

$$\times N\begin{pmatrix} m+p-1 & & m \\ & \cdots & \\ i_{m+p-1} & & i_m \end{pmatrix}. \quad (149)$$

For the formula of Eq. (149) to be identical to that obtained by Gould and Battle, Eq. (105), we must show that

$$P_{\alpha_{m+p}}(m+p)\cdot\left\langle \begin{matrix} p_{m+p}+\epsilon_{\alpha_{m+p}} \\ p_{m+p-1}+\delta_{i_{m+p-1}} \end{matrix} \middle\| \begin{matrix} p_{m+p} \\ p_{m+p-1} \end{matrix} \right\rangle$$

$$\cdot[R^{i_{m+p-1}}_{m+p-1}]^{-1/2}=V(\alpha_{m+p},i_{m+p-1}). \quad (150)$$

On the LHS the factor $P_{\alpha_{m+p}}(m+p)$ is given by Eqs. (141), the lexical adjoint-vector RWCs are listed in Appendix E as are the only two lexical RMEs. Note that these are shift dependent.

Proof of the above identity must be done on a case by case basis using lexicality considerations to match appropriate shifts and then to reduce the resulting expressions with the fundamental vector shifts.

If we rewrite Eq. (150) in the simplified form

$$L(\alpha_{m+p},i_{m+p-1})=V(\alpha_{m+p},i_{m+p-1}) \quad (151)$$

then we find that identity holds in all cases, viz.

$$\begin{aligned}
&L(+,1)=b_{m+p}+1 &&=V(+,1), &&\Delta p=(1,-1,1),\\
&L(+,1)=(-1)\cdot[(b_{m+p}+1)(b_{m+p}+3)]^{1/2} &&=V(+,1), &&\Delta p=(1,0,0),\\
&L(0,1)=(-1)\cdot\tfrac{1}{2}(b_{m+p}+2) &&=V(0,1), &&\Delta p=(0,1,0),\\
&L(0,1)=(-1)\cdot\tfrac{1}{2}b_{m+p} &&=V(0,1), &&\Delta p=(1,0,0),\\
&L(0,2)=\tfrac{1}{2}b_{m+p} &&=V(0,2), &&\Delta p=(1,-1,1),\\
&L(0,2)=\tfrac{1}{2}(b_{m+p}+2) &&=V(0,2), &&\Delta p=(1,0,0),\\
&L(-,2)=b_{m+p}+1 &&=V(-,2), &&\Delta p=(0,1,0),\\
&L(-,2)=(-1)\cdot[(b_{m+p}+1)(b_{m+p}-1)]^{1/2} &&=V(-,2), &&\Delta p=(1,0,0).
\end{aligned} \quad (152)$$

This establishes the complete equivalence of our del-operator ME formula with that of Gould and Battle.

VII. AN ALTERNATIVE APPROACH

In conclusion, we observe that there is yet another way to calculate the MEs of the del operator. Gould and Chandler[5] observed that there is a close relationship between this problem and the evaluation of the MEs of the two-body operator $X^i_j=E^i_{n+1}E^{n+1}_j$.

The starting point is the identity referred to in Sec. IV, Eq. (92). For convenience we reproduce it

$$\hat{\Delta}^i_j=\Delta(n+1)^i_j=E^i_{n+1}E^{n+1}_j+\Delta(n)^i_j+(\tfrac{1}{2}E^{n+1}_{n+1}-1)E^i_j. \quad (153)$$

We first note that the nonzero shift component of $\hat{\Delta}^i_j$ is simply

$$\Delta\begin{pmatrix} n \\ \pm \end{pmatrix}^i_j\equiv E^i_{n+1}E^{n+1}_i=X^i_j. \quad (154)$$

This is due to the fact that Δ^i_j and E^i_j are both zero shift in effect on the $U(n)$ Paldus label. Thus

$$\langle A|\Delta\binom{n}{\pm}^i_j|B\rangle=\langle A|X^i_j|B\rangle, \tag{155}$$

where $|A\rangle$ and $|B\rangle$ are GP basis states.

For the zero-shift component, we recall that

$$\Delta\binom{n}{0}^i_j=(\alpha_n+1)\cdot\Delta^i_j. \tag{156}$$

Now, the zero-shift component of Eq. (153) is given by

$$\Delta\binom{n}{0}^i_j=X\binom{n}{0}^i_j+\Delta^i_j+(\tfrac{1}{2}E^{n+1}_{n+1}-1)E^i_j. \tag{157}$$

On substituting for Δ^i_j from Eq. (156) we obtain

$$\left(\frac{\alpha_n}{\alpha_n+1}\right)\Delta\binom{n}{0}^i_j=X\binom{n}{0}^i_j+(\tfrac{1}{2}E^{n+1}_{n+1}-1)E^i_j. \tag{158}$$

Thus

$$\left(\frac{\alpha_n}{\alpha_n+1}\right)\cdot\langle P|\Delta\binom{n}{0}^i_j|Q\rangle$$

$$=\langle P|X\binom{n}{0}^i_j|Q\rangle+\langle P|(\tfrac{1}{2}E^{n+1}_{n+1}-1)E^i_j|Q\rangle, \tag{159}$$

where $|P\rangle$ and $|Q\rangle$ are once again GP basis states. In the above, the MEs of X^i_j and the $U(n)$ generators are known, Gould *et al.*[4,5,7]

VIII. CONCLUSION

In the above we have given an alternative derivation of the Gould–Battle formulas for the del-operator MEs based on ACCs and the Racah factorization lemma. This approach has the advantage that it extends to the general multishell case which we investigate in the forthcoming papers in this series.

The main purpose of this series of papers is to generalize the method outlined in Sec. VI above to obtain formulas for the MEs of the $U(2n)$ generators in a multishell spin–orbit basis. For this we require the two-shell adjoint WCs and adjoint-vector WCs and RWCs. These are evaluated in the second and third papers of the series together with the $U(2n)$ generator MEs in a two-shell spin–orbit basis. We also derive the adjoint reduced Wigner coefficients tabulated in Appendices D and E.

Finally, in paper four we derive the $U(2n)$ generator MEs in a multishell spin–orbit basis and demonstrate that in the GT limit they yield the correct one-shell formula as obtained by Gould and Battle[2] and rederived here.

APPENDIX A: ABBREVIATIONS

ACC	Adjoint coupling coefficient
ATO	Adjoint tensor operator
AVCC	Adjoint vector coupling coefficient
BR	Branching rules
CAS	Complete active space
CASCI	Complete active space configuration interaction
CGP	Composite Gelfand–Paldus
CI	Configuration interaction
CSF	Configuration state function
CVCC	Contragredient vector coupling coefficient
End(V)	The set of structure preserving mappings of a vector space V into itself, i.e., the set of endomorphisms
f.d.	Finite dimensional
GP	Gelfand–Paldus
GT	Gelfand–Tsetlin
irrep	Irreducible representation
MBPT	Many body perturbation theory
ME	Matrix element
MRCI	Multireference configuration interaction
RME	Reduced matrix element
RWC	Reduced Wigner coefficient
SCM	Shift component method
SO	Spin orbit
UGA	Unitary group approach
VCC	Vector coupling coefficient
WE	Wigner–Eckart

APPENDIX B: MATHEMATICAL LEMMA

Lemma 1

$\Delta(n)^i_j\equiv\Delta^i_j$ *is the unique—up to a multiple—adjoint tensor operator with the vanishing property that*

$$\Delta^i_j|\phi\rangle=0$$

if $|\phi\rangle$ *is any U(n) weight state with level i doubly occupied or level j unoccupied.*

Proof

Before outlining the proof of the lemma, we first recall the definition of a weight vector. If V is finite dimensional irrep of V then $v\in V$ is called a weight vector if v is an eigenvector of the n commuting operators E^i_i $(i=1,\ldots,n)$. That is, if

$$E^i_i v=\lambda_i v_i. \tag{B1}$$

The n-tuple $(\lambda_1,\lambda_2,\ldots,\lambda_n)$ is called the weight of the vector v. The natural lexicographical ordering on n-tuples allows the weights to be ordered.

In GP notation the irrep $V(a,b,c)=V(p)$ of $U(n)$ is uniquely labeled by the highest weight vector v with weight $(2^a,1^b,0^c)$, where $a+b+c=n$. For the irrep $V(2,2,1)\equiv V(2,2,1,1,0)$ of $V(5)$, where $a=2$, $b=2$, $c=1$ the state v defined by

$$v=\left|\begin{matrix}2&2&1&1&0\\&2&2&1&1\\&&2&2&1\\&&&2&2\\&&&&2\end{matrix}\right\rangle \tag{B2}$$

is called a maximal weight state. Clearly the weight of v is given by the 5-tuple (2,2,1,1,0), giving in this case the orbital occupancy of each level.

We divide the proof of the lemma into two parts:

(i) Proof of the vanishing property.
(ii) Proof of uniqueness.

Proof of the vanishing property

Let $|\phi\rangle$ be *any* weight state with orbital i doubly occupied or level j unoccupied. First suppose $i=j$. In this case

$$\Delta^i_i|\phi\rangle=(E^2)^i_i|\phi\rangle+(N/2-n-2)E^i_i|\phi\rangle$$
$$=(E^2)^i_i|\phi\rangle+(N-2n-4)|\phi\rangle. \qquad (B3)$$

Now,

$$(E^2)^i_i|\phi\rangle=\sum_j^{\text{unocc.}} E^i_jE^j_i|\phi\rangle+\sum_j^{\text{sing.occ.}} E^i_jE^j_i|\phi\rangle+\sum_j^{\text{doub.occ.}} E^i_jE^j_i|\phi\rangle$$
$$=\sum_j^{\text{unnocc.}}(\delta^j_jE^i_i-E^j_j+E^j_iE^i_j)|\phi\rangle+\sum_j^{\text{sing.occ.}}(\delta^j_jE^i_i-E^j_j+E^j_iE^i_j)|\phi\rangle+(E^i_i)^2|\phi\rangle$$
$$=\left[2\sum_j^{\text{unocc.}}+2\sum_j^{\text{sing.occ.}}-\sum_j^{\text{sing.occ.}}(E^j_j+4)\right]|\phi\rangle$$
$$=\left[2\sum_j^{\text{unocc.}}+\sum_j^{\text{sing.occ.}}+4\right]|\phi\rangle$$
$$=\left[2n-2\sum_j^{\text{doub.occ.}}-2\sum_j^{\text{sing.occ.}}+\sum_j^{\text{sing.occ.}}+4\right]|\phi\rangle$$
$$=\left[2n-\sum_{i=1}^n E^i_i+4\right]|\phi\rangle=(2n-N+4)|\phi\rangle. \qquad (B4)$$

Upon substitution of Eq. (B4) into Eq. (B3) we see that

$$\Delta^i_i|\phi\rangle=0, \qquad (B5)$$

as required.

Similarly, if j is unoccupied we have

$$\Delta^i_j|\phi\rangle=(E^2)^i_j|\phi\rangle+(N/2-n-2)E^i_j|\phi\rangle$$
$$=\sum_{k=1}^n E^i_kE^k_j|\phi\rangle+(N/2-n-2)E^i_j|\phi\rangle=0, \qquad (B6)$$

since $E^i_j|\phi\rangle=0$ whenever orbital j is unoccupied. This establishes the first part of the lemma.

Proof of uniqueness

To show uniqueness we need only work with a *maximal* state (see above).

Now suppose that

$$D^i_j=A(E^2)^i_j+BE^i_j+C\delta^i_j \qquad (B7)$$

is any $U(n)$ ATO with the stated vanishing property. In Eq. (B7) above A, B, and C are $U(n)$ invariants. The form of D^i_j follows the general theory of ATOs,[13] referred to in Sec. III, which states that any zero-shift $U(n)$ ATO can be expressed as a combination of the three independent (zero-shift) operators $(E^2)^i_j$, E^i_j, and δ^i_j.

We now show that D^i_j is a multiple of Δ^i_j.

(i) Let $|\phi\rangle$ be a $U(n)$ maximal state with orbital j unoccupied and take the ME of D^i_j in Eq. (B7) with $|\phi\rangle$, viz.

$$\langle\phi|D^i_j|\phi\rangle=A\langle\phi|(E^2)^i_j|\phi\rangle+B\langle\phi|E^i_j|\phi\rangle+C\langle\phi|\delta^i_j|\phi\rangle. \qquad (B8)$$

We have immediately, by the initial assumption, that the LHS of Eq. (B8) vanishes. If we now look at the special case $i=j$ and use the fact that[1]

$$(E^2)^j_j|\phi\rangle=0 \quad \text{and} \quad E^i_j|\phi\rangle=0 \qquad (B9)$$

then we have that $C=0$ so that Eq. (B8) becomes

$$D^i_j=A(E^2)^i_j+BE^i_j. \qquad (B10)$$

(ii) Now let $|\psi\rangle$ be a maximal $U(n)$ state with level i doubly occupied and, as before, take the ME of D^i_j in Eq. (B10) with $|\psi\rangle$. We obtain

$$\langle\psi|D^i_j|\psi\rangle=\langle\psi|(E^2)^i_j|\psi\rangle+B\langle\psi|E^i_j|\psi\rangle. \qquad (B11)$$

Once again the LHS of Eq. (B11) vanishes and using the results[1]

$$(E^2)^i_i|\psi\rangle=-2\gamma|\psi\rangle \quad \text{and} \quad E^i_i|\psi\rangle=2|\psi\rangle, \qquad (B12)$$

we see that

$$0=-2\gamma A+2B. \qquad (B13)$$

That is $B=\gamma A$ so that Eq. (B10) becomes

$$D^i_j=A\cdot\{(E+\gamma)E^i_j\} \qquad (B14)$$

or

$$D^i_j=A\cdot\Delta^i_j. \qquad (B15)$$

This establishes that any zero-shift ATO, D^i_j, with the vanishing property is a multiple of Δ^i_j, that is to say, the del operator is unique.

This completes the proof of lemma 1.

Lemma 2

$$\hat{\Delta}^i_j=\Delta(n+1)^i_j=\Delta(n)^i_j+X(n)^i_j+(\tfrac{1}{2}E^{n+1}_{n+1}-1)E^i_j, \qquad (B16)$$

where $X(n)^i_j=E^i_{n+1}E^{n+1}_j$ *and* $1\leq i,j\leq n$.

Proof

By definition

$$\Delta(n+1)^i_j=\hat{E}(\hat{E}+\gamma_{n+1})^i_j=(\hat{E}^2)^i_j+\gamma_{n+1}\hat{E}^i_j, \qquad (B17)$$

where $\hat{E}^k_l=E(n+1)^k_l$, $(1\leq k,l\leq n+1)$. Now from Gould and Battle[2] we have

$$\gamma_{n+1}=\hat{N}/2-\hat{n}-2$$
$$=\tfrac{1}{2}\sum_{k=1}^{n+1} E_k^k-(n+1)-2$$
$$=(\tfrac{1}{2}E_{n+1}^{n+1}-1)+\tfrac{1}{2}\sum_{k=1}^{n} E_k^k-n-2$$
$$=(\tfrac{1}{2}E_{n+1}^{n+1}-1)+\gamma_n. \quad \text{(B18)}$$

We also use the fact that

$$\hat{E}_j^i\equiv E_j^i, \quad 1\leqslant i,j\leqslant n, \quad \text{(B19)}$$

and

$$(\hat{E}^2)_j^i=\sum_{k=1}^{n+1} E_k^i E_j^k=E_{n+1}^i E_j^{n+1}+\sum_{k=1}^{n} E_k^i E_j^k$$
$$=X(n)_j^i+(E^2)_j^i. \quad \text{(B20)}$$

After substitution of the above into Eq. (B17) we obtain the final result

$$\hat{\Delta}_j^i=\Delta(n)_j^i+X(n)_j^i+(\tfrac{1}{2}E_{n+1}^{n+1}-1)E_j^i. \quad \text{(B21)}$$

APPENDIX C: BASIC SEGMENTATION FORMULAS

$$T^r(-,-)=(-1)^{\Delta b_r}\left(\frac{b_r+1}{b_r-1}\right)^{\Delta b_r/2},$$

$$T^r(0,0)=\frac{1+b_r+\Delta_r}{1+b_r}=\alpha_{r-1}+1,$$

$$T^r(0,\pm)=\frac{1}{1+b_r},$$

$$T^r(+,+)=(-1)^{\Delta b_r}\left(\frac{b_r+3}{b_r+1}\right)^{\Delta b_r/2},$$

$$T^r(\pm,0)=2[(1+b_r)(1+b_r\pm 2)]^{-1/2},$$

$$T^{n+1}(\pm)=\frac{1}{1+b_{n+1}},$$

$$T^{n+1}(0)=\frac{1+b_{n+1}+\Delta b_{n+1}}{1+b_{n+1}}=\alpha_n+1,$$

$$T_m(\pm)=[(1+b_m)(1+b_m\pm 2)]^{1/2},$$

$$T_m(0)=-\frac{\Delta b_m}{2}(1+b_m+\Delta b_m)=\Delta(m)_{mm}.$$

APPENDIX D: TABLE OF ADJOINT REDUCED WIGNER COEFFICIENTS

$$\left\langle \begin{matrix} p_{n+1} \\ p_n \end{matrix} \middle\| \begin{matrix} p_{n+1} \\ p_n \end{matrix} \right\rangle=\frac{S(n)}{S(n+1)}\cdot(\alpha_n+1),$$

$$\left\langle \begin{matrix} p_{n+1} \\ p_n+\epsilon_+ \end{matrix} \middle\| \begin{matrix} p_{n+1} \\ p_n \end{matrix} \right\rangle=\frac{1}{S(n+1)}\cdot\left[\frac{(n+2-c_n)(b_n+1)(n+2-a_n)}{(2+b_n}\right]^{1/2},$$

$$\left\langle \begin{matrix} p_{n+1} \\ p_n+\epsilon_- \end{matrix} \middle\| \begin{matrix} p_{n+1} \\ p_n \end{matrix} \right\rangle=\frac{1}{S(n+1)}\cdot\left[\frac{(b_n+1)(a_n+1)(c_n+1)}{b_n}\right]^{1/2},$$

$$\left\langle \begin{matrix} p_{n+1}+\epsilon_+ \\ p_n \end{matrix} \middle\| \begin{matrix} p_{n+1} \\ p_n \end{matrix} \right\rangle=-\left[\frac{(b_n+2)}{(b_n+1)(n+2-c_n)(n+2-a_n)}\right]^{1/2},$$

$$\left\langle \begin{matrix} p_{n+1}+\epsilon_- \\ p_n \end{matrix} \middle\| \begin{matrix} p_{n+1} \\ p_n \end{matrix} \right\rangle=-\left[\frac{b_n}{(b_n+1)(1+a_n)(1+c_n)}\right]^{1/2},$$

$$\left\langle \begin{matrix} p_{n+1}+\epsilon_+ \\ p_n+\epsilon_+ \end{matrix} \middle\| \begin{matrix} p_{n+1} \\ p_n \end{matrix} \right\rangle=\left(\frac{n+2-a_n}{n+3-a_n}\right)^{1/2}, \quad \Delta_0=(0,0,1),$$

$$=(-1)\left[\frac{(b_n+1)(b_n+4)(n+2-c_n)(n+2-a_n)}{(b_n+2)(b_n+3)(n+3-a_n)(n+3-c_n)}\right]^{1/2},$$

$$\Delta_1=(0,1,0)$$
$$=(-1), \quad \Delta_2=(1,-1,1)$$
$$=\left(\frac{n+2-c_n}{n+3-c_n}\right)^{1/2}, \quad \Delta_3=(1,0,0)$$

$$\left\langle \begin{matrix} p_{n+1}+\epsilon_- \\ p_n+\epsilon_- \end{matrix} \middle\| \begin{matrix} p_{n+1} \\ p_n \end{matrix} \right\rangle=\left(\frac{1+c_n}{2+c_n}\right)^{1/2}, \quad \Delta_0=(0,0,1)$$
$$=(-1), \quad \Delta_1=(1,0,0),$$
$$=(-1)\left[\frac{(b_n-2)(1+a_n)(b_n+1)(1+c_n)}{(b_n-1)(a_n+2)(b_n)(c_n+2)}\right]^{1/2},$$

$$\Delta_2=(1,-1,1)$$
$$=\left(\frac{1+a_n}{2+a_n}\right)^{1/2}, \quad \Delta_3=(1,0,0).$$

Note that

$$\left\langle \begin{matrix} p_{n+1}+\epsilon_+ \\ p_n+\epsilon_- \end{matrix} \middle\| \begin{matrix} p_{n+1} \\ p_n \end{matrix} \right\rangle \quad \text{and} \quad \left\langle \begin{matrix} p_{n+1}+\epsilon_- \\ p_n+\epsilon_+ \end{matrix} \middle\| \begin{matrix} p_{n+1} \\ p_n \end{matrix} \right\rangle$$

are both nonlexical. And also, $S(n)=-2S_n(S_n+1)$; where $S_n=b_n/2$ and that $n=a_n+b_n+c_n$. For the shift dependent cases $\Delta_i\equiv\Delta p_{n+1}=p_{n+1}-p_n$ $(i=0,1,2,3)$.

APPENDIX E: TABLE OF ADJOINT-VECTOR REDUCED WIGNER COEFFICIENTS AND REDUCED MATRIX ELEMENTS

$$\left\langle \begin{matrix} p_{n+1}+\epsilon_+ \\ p_n+\delta_1 \end{matrix} \middle\| \begin{matrix} p_{n+1} \\ p_n \end{matrix} \right\rangle$$
$$=\begin{cases} (n+2-a_n)^{-1/2}, & \Delta_2 \\ -\left[\dfrac{(b_n+3)(n+2-c_n)}{(b_n+2)(n+2-a_n)(n+3-c_n)}\right]^{1/2}, & \Delta_3 \end{cases}$$

$$\left\langle \begin{matrix} p_{n+1} \\ p_n+\delta_1 \end{matrix} \middle\| \begin{matrix} p_{n+1} \\ p_n \end{matrix} \right\rangle$$
$$=\begin{cases} \dfrac{(b_n+3)}{2S(n+1)}\left[\dfrac{(b_n+1)(n+2-c_n)}{(b_n+2)}\right]^{1/2}, & \Delta_1 \\ \dfrac{b_n}{-2S(n+1)}(n+2-c_n)^{1/2}, & \Delta_3 \end{cases}$$

$$\left\langle \begin{matrix} p_{n+1} \\ p_n+\delta_2 \end{matrix} \middle\| \begin{matrix} p_{n+1} \\ p_n \end{matrix} \right\rangle = \begin{cases} \dfrac{(b_n-1)}{2S(n+1)} \left[\dfrac{(b_n+1)(a_n+1)}{b_n}\right]^{1/2}, & \Delta_2 \\ \dfrac{(b_n+2)}{2S(n+1)} (a_n+1)^{1/2}, & \Delta_3 \end{cases}$$

$$\left\langle \begin{matrix} p_{n+1}+\epsilon_- \\ p_n+\delta_2 \end{matrix} \middle\| \begin{matrix} p_{n+1} \\ p_n \end{matrix} \right\rangle = \begin{cases} (1+c_n)^{-1/2}, & \Delta_1 \\ -\left[\dfrac{(b_n-1)(1+a_n)}{b_n(a_n+2)(c_{n+1})}\right]^{1/2}, & \Delta_3 \end{cases}$$

$$\left\langle \begin{matrix} p_{n+1} \\ p_n+\delta_1 \end{matrix} \middle\| \psi \middle\| \begin{matrix} p_{n+1} \\ p_n \end{matrix} \right\rangle = [R_n^1]^{1/2} = \begin{cases} \dfrac{(n+2-c_n)(1+b_n)}{(2+b_n)}, & \Delta_1 \\ (n+2-c_n), & \Delta_3 \end{cases}$$

$$\left\langle \begin{matrix} p_{n+1} \\ p_n+\delta_2 \end{matrix} \middle\| \psi \middle\| \begin{matrix} p_{n+1} \\ p_n \end{matrix} \right\rangle = [R_n^2]^{1/2} = \begin{cases} \dfrac{(1+a_n)(1+b_n)}{b_n}, & \Delta_2 \\ (a_n+1), & \Delta_3 \end{cases}$$

where

$$\Delta_0=(0,0,1), \quad \Delta_1=(0,1,0),$$

$$\Delta_2=(1,-1,1), \quad \Delta_3=(1,0,0).$$

In all of the above $\Delta_i \equiv \Delta p_{n+1} = p_{n+1} - p_n$, $(i=0,1,2,3)$. The RMEs here were defined originally in Gould and Chandler[4] and formulas for them may be found in Gould and Chandler.[7]

Note added in proof. It has been implicitly understood in Appendixes D and E that if the total spin $S=\frac{1}{2}b_{n+1}$ at level $n+1$ is zero then all $U(n+1)$ zero-shift RWCs are zero [cf. remarks following Eqs. (8) and (89)].

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