Coulomb Energy Averaged over the $n\ell^N$-Atomic States with a Definite Spin

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Abstract

A purely group-theoretical approach (for which the symmetric group plays a central rôle), based upon the use of properties of fractional-parentage coefficients and isoscalar factors, is developed for the derivation of the Coulomb energy averaged over the states, with a definite spin, arising from an atomic configuration $n\ell^N$. 

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1. Introduction

In the framework of an alternative parametrisation for the theory of complex spectra [1-3], Kibler and Katriel [2] have conjectured a formula, viz,

\[ 2^{S+1} E_{av}[n\ell^N] = \frac{1}{2} N(N - 1)\Sigma + \frac{1}{2} \left[ \frac{N}{2} \left( \frac{N}{2} + 1 \right) - S(S + 1) \right] \Delta \]  

(1)
giving the average Coulomb energy \( E_{av}[n\ell^N] \) for the states, with a fixed value of the spin \( S \), which arise from an atomic configuration \( n\ell^N \) of \( N \) equivalent electrons in a shell \( n\ell \).

In Eq. (1), the parameters \( \Sigma \) and \( \Delta \) are defined through

\[ 1^{E_{av}[n\ell^2]} = \Sigma + \Delta \quad 3^{E_{av}[n\ell^2]} = \Sigma \quad (2) \]
corresponding to the singlet \( (S \equiv S_0 = 0) \) and triplet \( (S \equiv S_0 = 1) \) states for the case \( N = 2 \). A formula equivalent to (1) and (2) has been derived independently by using the method of moments [4].

It is the aim of this short paper to give a straightforward proof of (1) by making use of fractional-parentage coefficients and of an unusual sum rule satisfied by isoscalar factors for a chain of compact groups closely connected to the permutation group. Indeed, we work out (in Section 2) an extension of formula (1), valid in the general case of a spin-independent two-body interaction, and then specialise it to the case of the Coulomb interaction. Some concluding remarks are given in Section 3. The relevant material (Racah’s lemma and orthogonality-completeness property for Clebsch-Gordan coefficients or isoscalar factors) concerned with the group-theoretical approach of Section 2 is relegated in an appendix.

2. Average Energies
In the Russell-Saunders coupling (or \(LS\)-coupling), the state vectors of the electronic configuration \(n\ell N\) (for atoms and ions) may be written as

\[
\Psi \equiv |n\ell^N[f]\alpha LSM_L M_S). \tag{3}
\]

Most of the symbols in (3) have their usual meaning. Let us simply precise that \([f]\) stands for a Young pattern, with two columns (say of lengths \(\varphi_1\) and \(\varphi_2\) with \(\varphi_1 \geq \varphi_2\)), which characterises the orbital part of \(\Psi\). The spin part of \(\Psi\) may be described by the Young pattern \([\varphi] \equiv [\varphi_1 \varphi_2]\), with two rows of lengths \(\varphi_1\) and \(\varphi_2\), which turns out to be the transposed pattern \([\tilde{f}]\) of \([f]\). The pattern \([\varphi]\) is (unambiguously) connected with the number of electrons \(N\) and the total spin \(S\) via

\[
\varphi_1 + \varphi_2 = N \quad \varphi_1 - \varphi_2 = 2S. \tag{4}
\]

Finally, the symbol \(\alpha\) in (3) collectively denotes the remaining labels that are necessary for a single-valued enumeration of the (allowed) atomic state vectors of the configuration \(n\ell^N\).

The vectors \(\Psi\) are expressed in a basis adapted to the following chain of groups

\[
U_{4\ell+2} \supset U_{2\ell+1} (\rightarrow SO_3 \rightarrow SO_2) \otimes U_2 (\rightarrow SU_2 \rightarrow U_1). \tag{5}
\]

Each of the vectors (3) spans the \textit{antisymmetric} irreducible representation class (IRC) \(\{1^N\}\) of the unitary group \(U_{4\ell+2}\). The Young patterns \([f]\) and \([\varphi]\) characterise IRC’s of the groups \(U_{2\ell+1}\) and \(U_2\) for the orbital and spin parts of \(\Psi\), respectively. The quantum numbers \(L\) and \(S\) refer to IRC’s of the subgroups \(SO_3\) and \(SU_2\) of \(U_{2\ell+1}\) and \(U_2\), respectively. Finally, the projections \(M_L\) and \(M_S\) indicate in turn IRC’s of the subgroups \(SO_2\) and \(U_1\) of \(SO_3\) and \(SU_2\), respectively.
For a given configuration $n^\ell N$, the total number of the vectors (3) allowed by the Pauli principle coincides with the dimension of the IRC $\{1^N\}$ of the group $U_{4\ell+2}$. It is thus given by $\dim \{1^N\} = C_{4\ell+2}^N$ in terms of binomial coefficients. Among these $C_{4\ell+2}^N$ state vectors, we select the ones having a fixed value $S$ of the spin. Obviously, the number of vectors $\Psi$ with $S$ fixed is nothing but the product $N_f(2S + 1)$, where $N_f$ is the dimension of the IRC $[f]$ of $U_{2\ell+1}$ and $2S + 1$ may be visualised as the dimension $N_\varphi$ of the IRC $[\varphi]$ of $U_2$.

In the present paper, we are interested in the average energy

$$2S + 1 E_{\text{av}}[n^\ell N] = \frac{1}{N_f} \frac{1}{2S + 1} \sum_{\alpha L} \sum_{M_L} \sum_{M_S} (n^\ell N[f] \alpha LS M_L M_S | V | n^\ell N[f] \alpha LS M_L M_S)$$

where $V$ is a spin-independent two-body Hamiltonian

$$V = \sum_{j>i=1}^N V_{ij} \quad \text{with} \quad V_{ij} \equiv V_{ij}(r_{ij}). \quad (7)$$

Equation (6) gives the average of $V$ over the $N_f(2S + 1)$ state vectors (3) having a fixed spin $S$. In Eq. (7), the sum on $i$ and $j$ is to be extended over the $N(N - 1)/2$ two-electron interactions $V_{ij}$; furthermore, $V_{ij}$ depends only on the distance $r_{ij}$ between the electrons $i$ and $j$.

Invariance of $V$ under the rotation group $SO_3$ and the spin group $SU_2$ ensures that (6) can be rewritten as

$$2S + 1 E_{\text{av}}[n^\ell N] = \frac{1}{N_f} \sum_{\alpha L} (2L + 1) (n^\ell N \alpha LS | V | n^\ell N \alpha LS). \quad (8)$$

The next step is to introduce coefficients of fractional parentage in order to calculate
the matrix elements of $V$ in (8). This leads to [5]

$$
(n^L_\alpha LS|V|n^L_\alpha LS) = \frac{1}{2} N(N - 1) \sum_{\alpha' L' S'} \sum_{L_0 S_0}
(\ell^N_\alpha LS|\ell^{N-2}_\alpha \alpha' L'S', \ell^2 L_0 S_0)
$$

(9)

$$
(n^L_\alpha LS|V_1|n^L_\alpha LS) = \frac{1}{2} N(N - 1) \sum_{\alpha' L' S'} \sum_{L_0 S_0}
(\ell^{N-2}_\alpha \alpha' L'S', \ell^2 L_0 S_0|\ell^N_\alpha LS).
$$

Following Racah [6, 7] and Neudatchin and Smirnov [8], the two-particle coefficient of fractional parentage $(\ell^N \{|\ell^{N-2}, \ell^2\} \ell^N)^*$ in (9) can be developed as

$$
(\ell^N_\alpha LS|\ell^{N-2}_\alpha \alpha' L'S', \ell^2 L_0 S_0) = \sqrt{\frac{n_f n_{f'}}{n_f}}
$$

(10)

$$
(\ell^N[f]_\alpha L|\ell^{N-2}[f']_\alpha' L', \ell^2[f_0]_L S_0)
$$

$$
(s^N[\tilde{f}]_S|s^{N-2}[\tilde{f}']_S', s^2[\tilde{f}_0]_S S_0)
$$

where $s = 1/2$. In Eq. (10), the IRC’s $[f]$, $[f']$ and $[f_0]$ refer to the group $U_{2\ell+1}$. However, the symbols $n_f$, $n_{f'}$ and $n_{f_0}$ denote the dimensions of $[f]$, $[f']$ and $[f_0]$ as considered as IRC’s of the permutation groups $S_N$, $S_{N-2}$ and $S_2$, respectively. From a group-theoretical viewpoint, the fractional-parentage coefficient $(\ell^N_\alpha LS|\ell^{N-2}_\alpha \alpha' L'S', \ell^2 L_0 S_0)$ is identical to the isoscalar factor $(\{1^{N-2}\alpha' L'S' + \{1^2\}L_0 S_0|\{1^N\}_\alpha LS)$ for the chain $U_{4\ell+2} \supset SO_3 \otimes SU_2$ [7]. Equation (10) then corresponds to the factorization (see Refs. [6] and [7])

$$
(\{1^{N-2}\}_\alpha L'S' + \{1^2\}L_0 S_0|\{1^N\}_\alpha LS) = (\{1^{N-2}\}_\alpha L'S' + \{1^2\}L_0 S_0|\{1^N\}_\alpha LS)
$$

(11)

in terms of isoscalar factors for the chains $U_{4\ell+2} \supset U_{2\ell+1} \otimes U_2$, $U_{2\ell+1} \supset SO_3$ and
\[ U_2 \supset SU_2. \] Indeed, the correspondence between (10) and (11) yields

\[
(\{1^{N-2}\}[f'][\tilde{f}'] + \{1^2\}[f_0][\tilde{f}_0]\{1^N\}[f][\tilde{f}]) = \sqrt{\frac{n_f n_{f_0}}{n_f}}
\]

\[
([f']\alpha' L' + [f_0] L_0)[f] \alpha L = (\ell^N [f] \alpha L \{\ell^{N-2}[f']\alpha' L', \ell^2 [f_0] L_0 \}
\]

\[
([\tilde{f}'] S' + [\tilde{f}_0] S_0)[\tilde{f}] S = (s^N [\tilde{f}] S \{s^{N-2}[\tilde{f}'] S', s^2[\tilde{f}_0] S_0 \})
\]

In Eqs. (10) and (12), it is clear that \( n_{f_0} = 1 \) for both IRC’s \([f_0] = [2] \) and \([f_0] = [11] \) of \( S_2 \). In the case \([f_0] = [2] \) only even values \( L_0 = 0, 2, \cdots, 2\ell \) are admissible and \( S_0 = 0 \) while for \([f_0] = [11] \) we have \( L_0 = 1, 3, \cdots, 2\ell - 1 \) and \( S_0 = 1 \). Furthermore in view of (12), the spin part \((s^N [\tilde{f}] S \{s^{N-2}[\tilde{f}'] S', s^2[\tilde{f}_0] S_0 \})\) of the fractional-pararentage coefficient (10) is trivial in the atomic case since the isoscalar factor \(([[f'] S' + [\tilde{f}_0] S_0)[\tilde{f}] S)\) for the chain \( U_2 \supset SU_2 \) is unity for the allowed values of \( S' \) and \( S_0 \). By combining Eqs. (8)-(12), we obtain

\[
2^{S+1} E_{av}[n^{\ell N}] = \frac{1}{2} N(N - 1) \frac{1}{N_f} \sum_{\alpha L} \sum_{\alpha' L'} \sum_{S'} \sum_{L_0} \sum_{S_0} \frac{n_{f'}}{n_f} (2L + 1) (n\ell^2 L_0 S_0 |V_{12}| n\ell^2 L_0 S_0) |([f']\alpha' L' + [f_0] L_0)[f] \alpha L|^2.
\]

(13)

An important step is now to effectuate the summations over \( \alpha L \) and \( \alpha' L' \) in (13). This may be easily done by using the orthogonality-completeness relation (22) (see appendix) applied to the (orbital) isoscalar factors for the chain \( U_{2\ell+1} \supset SO_3 \). We thus get

\[
2^{S+1} E_{av}[n^{\ell N}] = \frac{1}{2} N(N - 1) \sum_{S'} \sum_{L_0} \sum_{S_0} \Delta([f][f'] \otimes [f_0]) \frac{n_{f'}}{n_f} \frac{1}{N_{f_0}} (2L_0 + 1) (n\ell^2 L_0 S_0 |V_{12}| n\ell^2 L_0 S_0).
\]

(14)

By introducing in (14) the average energy

\[
2^{S_0+1} E_{av}[n^{\ell^2}] = \frac{1}{N_{f_0}} \sum_{L_0} (2L_0 + 1) (n\ell^2 L_0 S_0 |V_{12}| n\ell^2 L_0 S_0)
\]

(15)
for the configuration \( n \ell^2 \), we finally arrive at

\[
2S^1E_{av}[n \ell^N] = \frac{1}{2} N(N - 1) \sum_{s' \text{ or } [f']} \sum_{s_0 \text{ or } [f_0]}^{2S_0^1} E_{av}[n \ell^2] \Delta([f][f'] \otimes [f_0]) \frac{n_{f'}}{n_f}.
\]

Equation (16) provides us with a closed form expression for the \( N \)-electron average energy \( 2S^1E_{av}[n \ell^N] \) as a function of the two-electron average energy \( 2S_0^1 E_{av}[n \ell^2] \) in the case of a general spin-independent two-body interaction.

At this stage, it is convenient to use the \((\Sigma, \Delta)\)-parametrization defined by (2). In this parametrization, the energy \( 2S^1E_{av}[n \ell^N] \) is a linear combination of the parameters \( \Sigma \) and \( \Delta \). The coefficient of \( \Sigma \) in this linear combination is clearly \((1/2)N(N - 1)\). Since the parameter \( \Delta \) appears only in the singlets of \( n \ell^2 \), the coefficient of \( \Delta \) in \( 2S_0^1 E_{av}[n \ell^N] \) is \((1/2)N(N - 1)(n_f/n_f)\), where \( n_f \) refers to the Young pattern \([\bar{f}]\) deduced from \([f]\) by omiting its first row. (The latter assertion follows from the fact that \( S_0 = 0 \) for the singlet states so that \( S' = S \) and thus the sum on \([f']\) in (16) reduces to the orbital Young pattern \([\bar{f}]\) associated to the spin Young pattern \([\bar{\varphi}] \equiv [\varphi_1 - 1, \varphi_2 - 1]\) for the spin \( S \) and the number of electrons \( N - 2 \).) Therefore, Eq. (16) may be written in the form

\[
2S_0^1E_{av}[n \ell^N] = \frac{1}{2} N(N - 1) \left( \Sigma + \frac{n_{\bar{f}}}{n_f} \Delta \right).
\]

The final step is to calculate the ratio \( n_{\bar{f}}/n_f \) of the dimensions of the IRC’s \([\bar{f}]\) (for \( S_{N-2} \)) and \([f]\) (for \( S_N \)). This may be achieved by using the well-known formula (see for instance Ref. [9]) for the IRC’s of the symmetric group. We thus get

\[
\frac{n_{\bar{f}}}{n_f} = \frac{\varphi_2(\varphi_1 + 1)}{N(N - 1)}.
\]

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The introduction of (4) and (18) into (17) leads to Eq. (1). This completes the proof of the formula (1) for \(2S+1E_{\text{av}}[n\ell\ell^N]\) in the general case of a spin-independent two-body Hamiltonian. If we are interested in the special case where \(V\) is the repulsive Coulomb interaction (i.e., \(V_{ij} = e^2/r_{ij}\)), it is then sufficient to replace the parameters \(\Sigma\) and \(\Delta\) in (1) by the appropriate linear combinations of the Slater-Condon-Shortley parameters \(F^{(k)}\).

3. Closing Remarks

The main results of this paper [namely, Eqs. (16) and (1)] have been derived in the framework of atomic spectroscopy. Let us mention that the result (1) is also of relevance in the spectroscopy of partly-filled shell ions in condensed matter as far as \(S\) may be assumed to be a good quantum number (an assumption that is reasonable for transition-metal ions in crystals). The analysis leading to the results (1) and (16) rests on the use of the (single-configuration) shell-model and on the assumption of a unique single-particle radial wave-function for all the terms that the atomic (or crystal-field) configuration gives rise to.

In the physical situation where \(S\) is a good quantum number (this situation occurs in some cases in atomic spectroscopy and in the spectroscopy of transition-metal ions in crystals), Eq. (1) provides us with an expression of the barycenter of the levels for a particular spin multiplicity; then, the average energy for a given \(S\) corresponds to an observable and contact with experiment may be established. Another interest of the sum rule (1), valid even when \(S\) is not a good quantum number, is to be found in the fact that (1) is useful for the purpose of checking matrix elements when diagonalising the matrix of \(V\) by means of an electronic computer. As a third interest of (1), it is to be mentioned that the average energy (1) may be used in the method of spectral
distributions (mainly developed in nuclear physics) for the reconstruction of the global
distribution of the energy levels with a fixed value of $S$.

Let us observe that formulas which parallel Eqs. (16) and (1) might be derived in
the shell model of nuclear physics. In this connection, we may think of obtaining
average energies for fixed spin $S$, or fixed isospin $T$, or fixed spin $S$ and isospin $T$.
However, a complication arises when replacing $N$ electrons on a $n\ell$-atomic shell by $N$
nucleons on a $n\ell$-nuclear shell because the isospin degree of freedom manifests itself by
the replacement of the trivial spin chain $(U_2)_S \supset (SU_2)_S$ by the nontrivial spin-isospin
chain $(U_4)_{ST} \supset (U_2 \to SU_2)_S \otimes (U_2 \to SU_2)_T$ (cf. Ref. [10]). In this respect, the use
of class-sum operators [11, 12] might be of central interest. We hope to return on this
extension to nuclear physics in a forthcoming paper.

Finally, it is to be emphasized that the unusual sum rule (22) of the appendix is
of pivotal importance in the derivation of Eqs. (16) and (1). This sum rule is based
on Eq. (19) and on the Racah lemma. It is interesting to mention that Eq. (19) also
plays a fundamental rôlé in the derivation of sum rules for the intensity of two-photon
transitions between Stark levels of a transition ion in a liquid or crystal environment
[13].

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Appendix: Sum Rules for Coupling Coefficients
Let $G$ be a finite or compact group. We use $g$ to denote an IRC of $G$ and $\mu$ to classify the rows and columns of a (standard) unitary matrix representation $D^g$ associated to $g$. In this notation, the Clebsch-Gordan coefficients of the group $G$ are written $(g_1g_2\mu_1\mu_2|g_1g_2bg\mu)$ where $b$ is an internal multiplicity label to be used when the Kronecker product $g_1 \otimes g_2$ contains the IRC $g$ several times. The Clebsch-Gordan coefficients of $G$ satisfy ordinary unitarity relations controled by summations on $\mu_1\mu_2$ or $bg\mu$ (see for example Refs. [14] and [15]). In addition, they satisfy the following sum rule (referred to as an orthogonality-completeness relation [15])

$$\sum_{\mu_1} \sum_{\mu} (g_1g_2\mu_1\mu_2|g_1g_2bg\mu)^* (g_1g_2'\mu_1\mu_2'|g_1g_2'b'g\mu) = \Delta(g|g_1 \otimes g_2) \delta(g_2', g_2) \delta(\mu_2', \mu_2) \delta(b', b) \frac{\dim g}{\dim g_2}$$

where $\Delta(g|g_1 \otimes g_2)$ is 1 or 0 according to as $g$ is contained in $g_1 \otimes g_2$ or not. In the special case where $G$ is the group $SU_2$ (or $SO_3$), there is no need for the label $b$ and we have $g \equiv j$ (or $\ell$) and $\mu \equiv m_j$ (or $m_\ell$); in this case, Eq. (19) is a simple rewriting of one of the two ordinary unitarity relations for the Clebsch-Gordan coefficients $(j_1j_2m_1m_2|j_1j_2jm)$ that follows by using the symmetry property of the latter coefficients under the interchange $j_2 \leftrightarrow j$.

Let us now consider a subgroup $H$ of $G$. The label $\mu$ may then be replaced by the triplet $ah\gamma$, where $h$ stands for an IRC of the group $H$, $\gamma$ for an index to characterise the rows and columns of a (standard) unitary matrix representation $D^h$ associated to $h$, and $a$ for an external multiplicity label to be used when the IRC $h$ of $H$ occurs several times in the reduction of the IRC $g$ of $G$. In the $G \supset H$ basis, Eq. (19) may be transcribed as

$$\sum_{a_1h_1\gamma_1} \sum_{ah\gamma} (g_1g_2a_1h_1\gamma_1a_2h_2\gamma_2|g_1g_2bgh\gamma)^* (g_1g_2'a_1h_1\gamma_1a_2'h_2'\gamma_2'|g_1g_2'b'gh\gamma) = \Delta(g|g_1 \otimes g_2) \delta(g_2', g_2) \delta(a_2', a_2) \delta(h_2', h_2) \delta(\gamma_2', \gamma_2) \delta(b', b) \frac{\dim g}{\dim g_2}$$

(20)
The orthogonality-completeness relation (20) can be rewritten in terms of isoscalar factors for the chain $G \supset H$. For this purpose, we use the Racah factorisation lemma [6]

$$
(g_1 g_2 a_1 h_1 \gamma_1 a_2 h_2 \gamma_2 | g_1 g_2 b g a h \gamma) = \sum_\beta (g_1 a_1 h_1 + g_2 a_2 h_2 | b g a \beta h) (h_1 h_2 \gamma_1 \gamma_2 | h_1 h_2 \beta h \gamma)
$$

(21)

that gives an expression of a Clebsch-Gordan coefficient of $G$, in a $G \supset H$ basis, as a linear combination of Clebsch-Gordan coefficients $(h_1 h_2 \gamma_1 \gamma_2 | h_1 h_2 \beta h \gamma)$ of $H$. The coefficients of this linear combination are the isoscalar factors $(g_1 a_1 h_1 + g_2 a_2 h_2 | b g a \beta h)$ for the chain $G \supset H$. (In Eq. (21), the label $\beta$ is a multiplicity label of type $b$.) Then, by applying twice (21) in (20) and by using (19) for the group $H$ in the so-obtained equation, we end up with

$$
\sum_{a_1 h_1} \sum_{a h} \sum_\beta \frac{\dim h_1}{\dim h_2} (g_1 a_1 h_1 + g_2 a_2 h_2 | b g a \beta h)^* (g_1 a_1 h_1 + g'_2 a'_2 h_2 | b' g a \beta h) = \Delta(g | g_1 \otimes g_2) \delta(g'_2, g_2) \delta(a'_2, a_2) \delta(b', b) \frac{\dim g}{\dim g_2}.
$$

(22)

Equation (22) constitutes an orthogonality-completeness relation for the isoscalar factors of the chain $G \supset H$. 
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