

Many Correlation Tables are Molien Sequences

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Using the Jordan-Schwinger form of the quantum angular momentum eigenstates, it is straightforward to define rotational correlation tables such that the columns are Molien sequences for finite rotational subgroup G . This realization gives a new and better means of calculation. Generalization to unitary symmetry $U(n)$ implies many more sequences, which determine degeneracy observables in the context of electronic, vibrational, and rotational motion. This leads us to discuss one physical significance of the Hilbert finite basis theorem.

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I. INTRODUCTION

The use of correlation tables to capture degeneracy of eigenstates due to subgroup structure $G \supset SG$ dates back to the early years of the new quantum mechanics [1]. Under any perturbation that breaks Hamiltonian symmetry from G to SG , elements of a correlation table, also called f -numbers, determine splitting of eigenstates. Level-splitting directly affects spectral measurements such as the intensity of dipole absorption whenever a laser irradiates a solid or molecule. Ease of accessibility gives the f -numbers importance as quantum observables.

As an example, consider quantum angular momentum. Spherical symmetry $SO(3)$ admits circular dihedral symmetry D_∞ as a subgroup, so we expect to find non-trivial degeneracy in the absence of symmetry-breaking perturbations. The well-known eigenstates have quantum numbers (j, m) , which admit the correlation of Table I.

Table I. $SO(3) \supset D_\infty$ Correlation.

$j \setminus m$	0	$\pm 1/2$	± 1	$\pm 3/2$	± 2	$\pm 5/2$	± 3	\dots
0	1	0	0	0	0	0	0	\dots
1/2	0	1	0	0	0	0	0	\dots
1	1	0	1	0	0	0	0	\dots
3/2	0	1	0	1	0	0	0	\dots
2	1	0	1	0	1	0	0	\dots
5/2	0	1	0	1	0	1	0	\dots
3	1	0	1	0	1	0	1	\dots
\vdots	\vdots	\vdots	\vdots	\vdots	\vdots	\vdots	\vdots	\ddots

The spectra of a rotating molecule follows from the specification of a Hamiltonian, $\Omega(\mathbf{J})$, a function of quantum mechanical angular momentum variables $\mathbf{J} = (J_x, J_y, J_z)$. In a semi-classical theory

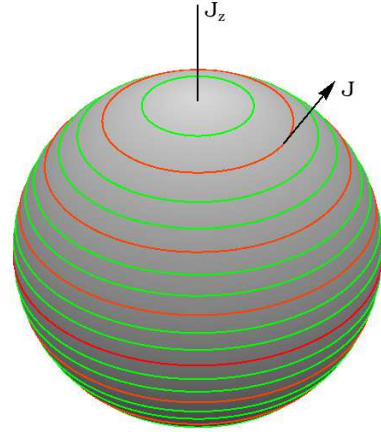


FIG. 1. Rotational Energy Surface. Weak perturbations do little to distort the shape of the RES. Trajectories at $j = 10$ are colored according to representations of D_3 symmetry: A_0 red, A_0/A_1 red-orange, E_1 green.

of rigid quantum rotations, the Hamiltonian determines a rotational energy surface (RES) where the \mathbf{J} vector moves, approximately, along quantized level sets [2]. Description of planar molecules involves a descent of symmetry

$$\Omega = \frac{\omega}{\hbar^2} \mathbf{J} \cdot \mathbf{J} + \frac{\xi_z}{\hbar^2} \mathbf{J} \cdot \mathbb{P}_z \cdot \mathbf{J} + \xi_\theta \theta(\mathbf{J}/\hbar), \quad (1)$$

where \mathbb{P}_z is a uniaxial projection matrix and θ is a higher-order polynomial function of the \mathbf{J} variables. The approximate symmetry of the atomic nuclei constrain the range of possibilities for θ . Fig. 1 depicts a spherical RES with slight perturbations.

Frequency hierarchy $\xi_\theta \ll \xi_z \ll \omega$ characterizes the simplest case where, to order ξ_z , the RES is an ellipsoid with just one circular cross section. Frequency levels split according to Table I. At $j = 3$, we have doublets corresponding to $m = \{\pm 1, \pm 2, \pm 3\}$ as well as one singlet $m = 0$. Including terms to order

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ξ_θ lifts some, possibly all, of the remaining degeneracy.

Calculations of rotational frequency levels and spectroscopic measurements of octahedral molecules at high- j reveal intriguing degeneracy sequences [3]. Considering a great number of excited states, it becomes possible to extend upon and improve the symmetry analysis available through the original crystal field theory. Harter explains how patterns of degeneracy follow the f -numbers of various correlations between spherical, octahedral, and cyclic symmetry groups [4].

The standard theory common between solid state physics and molecular physics [5] defines rotational correlations according to a character formula

$$CT(SO(3) \supset G) = \tilde{\chi} \cdot \chi^{-1}. \quad (2)$$

where χ and $\tilde{\chi}$ are matrices of characters, the traces of matrices that represent group actions. Each row of character table χ corresponds to an irreducible representation (irrep) of G , while each column corresponds to a conjugacy class of G . The rows and columns of $\tilde{\chi}$ correspond to irreps of $SO(3)$ and the conjugacy classes of G . The book *Principles of Symmetry, Dynamics, and Spectroscopy* [5] completely explains character theory, up to the derivation of rotational tables in Chapter 5.

General and important features of rotational correlation tables are known and utilized since 1929: The table rows have a periodic structure. Whole integer representations of angular momentum correlate to representations of the subgroup G while half-integer representations correlate only to representations of the double subgroup $2G$. According to these observations it should be possible to write the correlation tables as a set of generating functions.

Referencing various column sequences in the Online Encyclopedia of Integer Sequences (OEIS) [6], we find many suggestive connections between rotational correlation tables and various Molien series. Following the references through diversions into coding theory [7] and combinatorics [8], we make the realization that classical invariant theory also applies to quantum rotations and vibrations. The following presentation provides an alternative, or at least a supplement, to the standards currently available.

II. QUANTUM INVARIANT THEORY

In the history of science, the advent of quantum transformation theory follows after classical invariant theory [9]. The old invariant theory, in its general form, is founded upon famous theorems by Hilbert, Noether, and Molien. These century old

theorems have practical value in a wide range of applications [7],[8]. They apply to quantum mechanics wherever symmetry breaking lowers unitary to a finite subgroup. We show that classical and quantum transformation theories admit a natural and fruitful combination by considering polynomials of commuting variables, the familiar quantum harmonic oscillator raising operators a_i^\dagger .

The $SU(2) \sim SO(3)$ double cover requires the use of double groups. This critical insight allows us to improve and extend the notion of a rotational correlation table. By our definitions, Molien's theorem implies the following corollaries

- **Corollary 1.** The f -numbers of rotational correlation table $CT(U(2) \supset 2G : \Gamma_{1/2})$ are determined entirely by the Molien equation when $\Gamma_{1/2}$ is a $j = 1/2$ representation of G .
- **Corollary 2.** For any n -dimensional irreducible representation Γ of finite symmetry group G , the f -numbers of correlation table $CT(U(n) \supset G : \Gamma)$ are determined entirely by the Molien equation.

In stating these corollaries we use a naming convention slightly different from Eq. 2, which is required in a wider context that applies throughout the Born-Oppenheimer hierarchy; to the electronic, vibrational, and rotational motion of molecules and solids.

A. Molien Equation

Molien's equation,

$$gf(\Gamma_x, \Gamma_y, \lambda) = \frac{1}{|G|} \sum_{A_i \in \Gamma_x} \frac{\chi^*(\Gamma_y, i)}{\text{Det}[\mathbb{I} - A_i \lambda]} \quad (3)$$

$$= f_0(\Gamma_y) + f_1(\Gamma_y)\lambda + f_2(\Gamma_y)\lambda^2 + \dots$$

is a clever utility that automates the analysis of induced representations, thus enabling the counting of invariants and covariants.

Representation $\{A_1, A_2, \dots, A_g\} \in \Gamma_x$ is a group of $m \times m$ dimensional matrices A_i that act linearly on polynomial variables $\mathbf{x} = (x_1, x_2, \dots, x_m)$. This one representation of G determines an infinite set of representations $\{A_1^n, A_2^n, \dots, A_g^n\} \in \Gamma_x^n$. Each representation Γ_x^n acts linearly on a complete set of polynomials of homogeneous order n , $\mathbf{x}^n = (x_1^n, x_1^{n-1}x_2, \dots, x_2^n, x_2^{n-1}x_3, \dots, x_m^n)$. Notice that a transformation of the \mathbf{x} variables always causes an element of \mathbf{x}^n to transform into a linear combination of all elements of \mathbf{x}^n .

A subspace $\mathbf{s}^n = \mathbb{P}_s^n \mathbf{x}^n \in \mathbf{x}^n$ is said to transform covariantly to irrep Γ_y whenever the Γ_y projector

acts as identity

$$\mathbf{s}^n = \mathbb{P}_{\Gamma_y}^n \mathbf{s}^n. \quad (4)$$

If Γ_y is the trivial irrep with all characters equal to one, then \mathbf{s}^n is also said to transform invariantly. This definition gives the projectors $\mathbb{P}_{\Gamma_y}^n$ central importance in any attempt to enumerate sequences of covariants.

To define the idempotent projectors [5], we need the irreducible characters $\chi(\Gamma_y, i)$ of the i^{th} element of group G . These characters are listed by conjugacy class in the character tables. Then we have,

$$\mathbb{P}_{\Gamma_y}^n = \frac{1}{|G|} \sum_{A_i \in \Gamma_x} \chi^*(\Gamma_y, i) A_i^n, \quad (5)$$

where the $*$ indicates complex conjugation. Every polynomial of homogeneous order n transforms covariantly to some irrep according to the decomposition of the identity operator

$$\mathbb{I}^n = \sum_y \chi(\Gamma_y, 1) \mathbb{P}_{\Gamma_y}^n, \quad (6)$$

where $i = 1$ is assumed to index the identity operation $\mathbb{I} = A_1$, and the sum is taken over all irreps.

Clearly the task of counting the number of polynomial subspaces with some particular transformation property reduces to computation of projector traces

$$f_n(\Gamma_y) = \text{Tr}[\mathbb{P}_{\Gamma_y}^n], \quad (7)$$

where by Eq. 6, $f_n(\Gamma_y)$ counts each $\chi(\Gamma_y, 1)$ -dimensional multiplet exactly once. According to properties of the trace function, $\text{Tr}[\mathbb{P}_{\Gamma_y}^n]$ is determined from the quantities $\text{Tr}[A_i^n]$ alone.

Fortunately, Molien notices that

$$\frac{1}{d(\Gamma_x, i, \lambda)} = \frac{1}{\text{Det}[\mathbb{I} - A_i \lambda]} = \sum_n \text{Tr}[A_i^n] \lambda^n, \quad (8)$$

thus it becomes unnecessary to calculate induced representations explicitly. Finally, the proof of the covariant Molien theorem rests upon the same crux as the invariant theorem nicely re-proven by Sloane [7].

- **Theorem 1. (Molien)** Coefficient $f_n(\Gamma_y)$ of λ^n in the power series expansion of the generating function $gf(\Gamma_x, \Gamma_y, \lambda)$ gives the number of Γ_y -covariants of homogeneous order n in variables $\mathbf{x} = (x_1, x_2, \dots, x_m)$ that transform according to a representation Γ_x of group G .

We apply this theorem in a physical context.

B. The Pseudotop Analogy

The pseudotop analogy[10] occurs in quantum mechanics wherever a quantum system with fixed spatial orientation transforms according to a rotational algebra. Numerous resources [5],[11],[12],[13] discuss this analogy in the context of the two-dimensional isotropic quantum harmonic oscillator. Here the pseudotop analogy takes a most simple form, which is both a sincere curiosity and a useful aid to practical calculations. Recall the Jordan-Schwinger form for the eigenstates of angular momentum

$$|j, m\rangle \propto (a_1^\dagger)^{j+m} (a_2^\dagger)^{j-m} |0, 0\rangle, \quad (9)$$

where $(j, m) = \frac{1}{2}(n_1 + n_2, n_1 - n_2)$ provides a relation between rotational quantum numbers (j, m) and vibrational quantum numbers (n_1, n_2) .

Polynomials of the commuting raising operators $\boldsymbol{\eta} = (a_1^\dagger, a_2^\dagger)$ determine all representations of continuous group $SU(2) \sim SO(3)$. As in section IIa we define induced spaces $\boldsymbol{\eta}^{2j}$ where polynomials of the $\boldsymbol{\eta}$ variables have homogeneous order $2j$. All polynomials transform invariantly by identity symmetry, so the trivial Molien equation in two-dimensions gives the generating function for the total degeneracy

$$gf(\{\begin{pmatrix} 1 & 0 \\ 0 & 1 \end{pmatrix}\}, \{(1)\}, \lambda) = 1/(1 - \lambda)^2 \quad (10) \\ = 1 + 2x + 3x^2 + 4x^3 + 5x^4 \dots$$

The series coefficients equal the sum of columns in Table I above.

In a situation where finite rotational symmetry G constrains a Hamiltonian with approximate spherical symmetry $SU(2) \sim SO(3)$, we would also like to impose the constraints of G onto the eigenstates. This is usually done using projectors $\mathbb{P}_{\Gamma_y}^n$, but classical invariant theory provides an alternative approach that we like to explore.

Our conventions present the only difficulty in applying Theorem 1 directly. Usually we start with a real-space, $j = 1$ representation Γ_1 , but $\boldsymbol{\eta}$ transforms according to the $j = 1/2$ representation $\Gamma_{1/2}$. To perform reverse-induction, define

$$\boldsymbol{\eta}_i^2 = \boldsymbol{\eta} \cdot g_i \cdot \boldsymbol{\eta}, \quad (11)$$

where $\mathbf{g} = \frac{1}{2}(\mathbb{I} + \sigma_3, \sqrt{2} \sigma_1, \mathbb{I} - \sigma_3)$ with σ_i the standard Pauli matrices.

The matrix/vector \mathbf{g} transforms according to either

$$\mathbf{g} \longrightarrow \mathbf{g}' = \mathcal{D}_{\alpha, \beta, \gamma}^1 \cdot \mathbf{g}, \quad (12a)$$

$$g_i \longrightarrow g'_i = (\mathcal{D}_{\alpha, \beta, \gamma}^{1/2})^T \cdot g_i \cdot \mathcal{D}_{\alpha, \beta, \gamma}^{1/2}, \quad (12b)$$

where $\mathcal{D}_{\alpha, \beta, \gamma}^j$ is a j -irrep rotation matrix with Euler angles (α, β, γ) [5].

Equation 12 provides an equivalence between vector rotation and matrix conjugation that allows the determination of $\Gamma_{1/2}$ from Γ_1 . A difficulty arises because the similarity transform allows $-\mathbb{I}$ to act as identity. This opens up the possibility that Γ_1 is not a faithful representation of the group G represented by $\Gamma_{1/2}$, that $\Gamma_{1/2}$ may contain more distinct elements than Γ_1 .

The general form of $\mathcal{D}_{\alpha,\beta,\gamma}^{1/2}$ is

$$\begin{aligned} \mathcal{D}_{\alpha,\beta,\gamma}^{1/2} &= e^{-i\frac{\alpha}{2}\sigma_3} e^{-i\frac{\beta}{2}\sigma_2} e^{-i\frac{\gamma}{2}\sigma_3} \quad (13) \\ &= \begin{pmatrix} e^{-i(\alpha+\gamma)/2} \cos \beta/2 & -e^{-i(\alpha-\gamma)/2} \sin \beta/2 \\ e^{i(\alpha-\gamma)/2} \sin \beta/2 & e^{i(\alpha+\gamma)/2} \cos \beta/2 \end{pmatrix}. \end{aligned}$$

The factor of 2 attached to all angles in Eq. 13 causes all 2π rotations to have a representation

$$\mathcal{D}_{2\pi}^{1/2} = -\mathbb{I}, \quad (14)$$

while, intuitively, a 2π rotation in Γ_1 must act as identity. This discrepancy requires that the $\Gamma_{1/2}$ representation contains twice as many elements as the Γ_1 representation and explains the locution that ' $SU(2)$ is the double cover of $SO(3)$ '. We agree with Klein [14] that the $\Gamma_{1/2}$ representation of G is actually a representation of the double group $2G$.

$\Gamma_{1/2}$ takes a special place among all $2G$ irreps because of spin and the pseudotop analogy. By Eq. 11, $\Gamma_{1/2}$ also transforms $\boldsymbol{\eta}$, so it is exactly the representation we need to use the tools of classical invariant theory in an analysis of quantum rotations. Setting $\Gamma_x = \Gamma_{1/2}$ in the Molien equation, we have

$$CT(U(2) \supset 2G : \Gamma_{1/2}) = gf(\Gamma_{1/2}, \Gamma_y, \lambda), \quad (15)$$

where each Γ_y of group $2G$ determines a column of the correlation table. We assert that corollary 1 is proven by definition, but it is also possible to prove equivalence between the character formula and the Molien equation by comparing how each method treats the decomposition of the $Tr[\mathbb{P}_{\Gamma_y}^n]$.

1. Examples

Using Eqs. 12 we generate $\Gamma_{1/2}$ from Γ_1 for each of the following groups: Triangular Dihedral (D_3), Octahedral (O), and Icosahedral (A_5). From the $\Gamma_{1/2}$ representations we calculate correlation tables $CT(U(2) \supset 2G : \Gamma_{1/2})$ according to Molien's Eq. 2. These tables, available on-line [15], are the same as typically obtained [1],[4],[16],[17] by character formula Eq. 2, up to an arbitrary g or u label. The on-line supplement also contains an incomplete cross-reference of the column sequences with entries in the OEIS [6]. Some of the sequences computed in this investigation seem to be missing from the extensive records.

C. Generalization

Corollary 2 generalizes corollary 1 by removing the restriction $\Gamma_x = \Gamma_{1/2}$. This is an important move because quantum mechanics involves many symmetries other than $SU(2) \sim SO(3)$. In a sufficiently general setting we have an n -dimensional representation Γ_n of group G which is also a subgroup of $U(n)$. The representation generators $\boldsymbol{\eta}_n = (a_1^\dagger, a_2^\dagger, \dots, a_n^\dagger)$ are the ladder operators of the n -dimensional isotropic quantum harmonic oscillator. These commuting variables we again subject to the constraints of symmetry.

The molecular physics literature already contains numerous examples in the general setting, including some investigation of the Molien function[18]. We review a few articles and provide Molien generating functions that help to explain symmetry classification of excited vibrational states.

1. Examples

Level correlations in [19] are given by

$$\begin{aligned} SF_6, UF_6 &: CT(U(3) \supset O : T_1), \\ SiF_4 &: CT(U(3) \supset O : T_2). \end{aligned}$$

Tables available on-line [15] give f -numbers consistent with the published levels.

Level correlations in [20],[21] are slightly more complicated, but nevertheless yield to a Molien analysis. The separate ladders obey

$$\begin{aligned} |\nu_1, 0, 0\rangle &: CT(U(1) \supset D_3 : A_1), \\ |0, \nu_2, 0\rangle &: CT(U(1) \supset D_3 : A_1), \\ |0, 0, \nu_3\rangle &: CT(U(2) \supset D_3 : E); \\ |\nu_1, 0, 0\rangle &: CT(U(1) \supset O : A_{1g}), \\ |0, \nu_2, 0\rangle &: CT(U(2) \supset O : E_g), \\ |0, 0, \nu_3\rangle &: CT(U(3) \supset O : F_{1u}). \end{aligned}$$

Considering combinatorial properties of the Molien equation it is possible to join separate correlation tables into one table that gives correlations for combined states $|\nu_1, \nu_2, \nu_3\rangle$. In these tables the f -numbers are coefficients of a power series expansion in three formal variables $\lambda_1, \lambda_2, \lambda_3$. We write the combined-ladder correlation tables as

$$\begin{aligned} |\nu_1, \nu_1, \nu_3\rangle &: CT(U(1) \oplus U(1) \oplus U(2) \supset \\ & \quad D_3 : A_1 \oplus A_2 \oplus E). \\ |\nu_1, \nu_1, \nu_3\rangle &: CT(U(1) \oplus U(2) \oplus U(3) \supset \\ & \quad O : A_{1g} \oplus E_g \oplus T_{1u}). \end{aligned}$$

All level degeneracies calculated using the Molien function agree with published results.

D. Finite Reduction of Eigenspaces

The Molien equation allows us to count the number of excited states transforming covariantly to Γ_y but does not exactly provide a means for enumerating the states themselves. We could construct the projectors $\mathbb{P}_{\Gamma_y}^n$ by induction. Other authors [21], [22] call the brute force method difficult, and seek out new, creative approaches. According to the Hilbert finite basis theorem, invariant theory provides another alternative.

Preexisting theorems (Cf. [8] Thm. 1.3, 3.10) guarantee the existence of a simple procedure that generates a complete span of each Γ_y eigenspace. The algorithm depends only on the input of a finite set of polynomials, which belong to either one of two classes [7],[8]. Surmounting the difficulties of troublesome syzygies, we can arrive at a succinct and sophisticated specification of the eigenspaces, which allows us to classify excited states of a perturbed oscillator.

1. Example

Examining the $2D_3 : E_{1/2}$ Molien functions and various idempotent projectors for $2j = 1, 2, 3, 4, 5, 6$, we determine a finite set of polynomial invariants and covariants. From these, we write a basis of rotational states

$$|j, m, \chi\rangle \propto (a_1^\dagger a_2^\dagger)^{(j-m)} ((a_1^\dagger)^{2m} + \chi (a_2^\dagger)^{2m}) |0\rangle, \quad (16)$$

with $j \geq m \geq 0$ and $\chi = \pm 1$ or $\pm i$. Apparently these are a complete, orthogonal set of J_z^2 eigenfunctions. This basis also divides nicely into classes that transform covariantly to the representations of $2D_3$. Table II gives the relation between quantum numbers and symmetry labels.

Γ_y	$(j - m)$	$2m \% 6$	χ
A_0	Even	0	+1
	Odd	0	-1
	Even	$m = 0$	0
A_1	Even	0	-1
	Odd	0	+1
	Odd	$m = 0$	0
E_1	.	2, 4	± 1
$E_{3/2}^L$	Even	3	$+i$
	Odd	3	$-i$
$E_{3/2}^R$	Even	3	$-i$
	Odd	3	$+i$
$E_{1/2}$.	1, 5	± 1

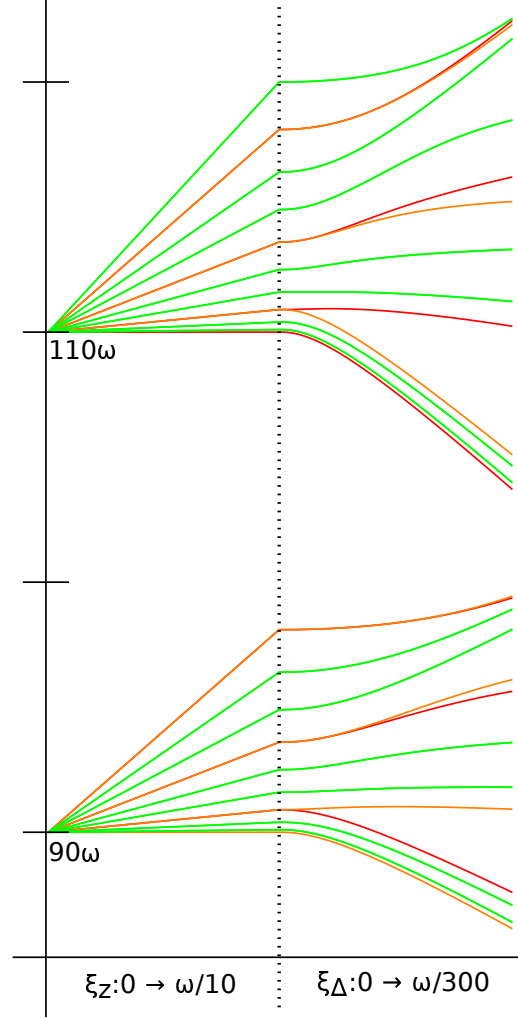


FIG. 2. Rotational Levels. The $j = 9, 10$ rotational levels are colored according to symmetry type A_0 red, A_1 orange, E_1 green. From left to right, a J_z^2 perturbation of strength ξ_z is applied, followed by a D_3 perturbation of strength ξ_Δ . The D_3 perturbation causes the A_0/A_1 doublets to split.

This table is completely consistent with table $CT(U(2) \supset 2D_3 : E_{1/2})$ available on-line [15]. We use table II and Eq. 16 to label level-splitting diagrams such as Fig. 2.

III. CONCLUSION

Direct involvement of the unitary creation operators clarifies the application of classical invariant theory to quantum mechanics. These polynomial variables often occur in molecular physics, as the generators of quantized rotations or vibrations. We

give a unified perspective that views electronic, rotational, and vibrational multiplet counting as equivalent tasks. The correlation tables defined herein apply immediately to any molecule or solid with dihedral, tetrahedral, octahedral, or icosahedral symmetry.

As with any of the best mathematical theories, the range of applications for classical invariant theory is truly interdisciplinary. If we confine ourselves to studying physics, we still expect to find connections wherever unitary symmetry occurs. One as-yet unexplored possibility exists in quantum optics [23], where correlation techniques should apply to solutions of the plane-wave Helmholtz equation.

This letter focuses on the simple Molien equation, which is only a small part of the complete invariant theory. As this research continues, we confront the exciting possibilities and the difficult challenges (syzygies) that arise in a closer examination of the Hilbert finite basis theorem. Immediately we find a useful equivalence between polynomial rings and irreducible eigenspaces. Explicit construction and manipulation of creation-operator polynomials may lead to new discovery in physics.

Invariant theory is an important chapter in the

history of science, because it already contains many notions essential to the foundations of modern physics. Clebsch and Gordan obtained early results regarding polynomials generated by two variables. In the generalization to an arbitrary number of polynomial generators, Hilbert affected a change of paradigm that brought new methods. We present one of those methods here, and suggest more investigation. Our hope is that the historical drama, taken alongside the content of this article, will give physicists vital motivation to seriously consider new applications of XIX century techniques.

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