The Master Thesis of Moshé Flato

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to my Master and Friend Moshé Flato

Abstract
The genesis and impact of the M.Sc. thesis by Moshé Flato is analysed. In this connection, the fruitful passage of Moshé in Lyon, capitale mondiale de la gastronomie, is evoked. Finally, some basic elements for a model in crystal-field theory are given as an important step on the way opened by Moshé.

Key words: finite subgroups of SU(2), Wigner-Racah algebra for finite and compact groups, crystal-field theory, molecular and condensed matter spectroscopy.

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Moshé Flato prepared his M.Sc. thesis under the guidance of Giulio Racah at the Physics Department of the Hebrew University in Jerusalem at the end of the fifties [1]. The final examination took place in 1960. The genesis of the thesis can be described as follows.

From 1942 to 1949, Racah published a series of four major papers on new algebraic and group theoretical methods for the analysis of complex spectra in atomic and nuclear spectroscopy [2-5]. In particular, he developed the concepts of irreducible tensor operator, coefficient of fractional parentage (a concept which goes back to Goudsmit and Bacher) and seniority number. Furthermore, he showed how the use of a chain of continuous groups ending with \( SO(3) \supset SO(2) \) makes it possible to calculate the coefficients of fractional parentage and to classify the state vectors originating from an \( \ell^N \) configuration in spherical symmetry. In 1956, he became very interested in crystal-field theory (a theory for partly-filled shell ions embedded in crystals, solutions or complexes) on the occasion of discussions and seminars with Wily Low from the Microwave Division of the Department of Physics of the Hebrew University [6]. Crystal-field theory, also referred to as ligand-field theory, may be considered as a branch of quasi-atomic physics in the sense that this theory is concerned with the electronic properties of a central ion (with a configuration of type \( \ell^N \) where \( \ell = d \) or \( f \)) in interaction with its immediate surrounding. Racah realized that the simultaneous consideration of finite groups (for describing the ion in its environment) and continuous groups (for labelling the Russell-Saunders terms \([S, L]\) or multiplets \([J]\) of the ion) can be very fruitful in crystal-field theory. W. Low and G. Racah suggested to two students, Miss G. Schoenfeld and Mr. G. Rosengarten, to calculate the Coulomb, spin-orbit and crystal-field matrices for \( d^N \) ions in cubical symmetries. The cubical symmetry is often an idealized (or first-order) symmetry so that G. Racah asked Mr. M. Flato to reconsider the problem of \( d^N \) ions in finite symmetry and to treat the case of trigonal and tetragonal symmetries which are of interest in absorption spectroscopy and in electron spin resonance spectroscopy.

Schoenfeld calculated the matrices for the \( d^2 \) and \( d^3 \) configurations in cubical symmetry (symmetry group \( G = O \)) and Rosengarten obtained the matrices for the \( d^4 \) and \( d^5 \) configurations in cubical symmetry too. Flato dealt with the case where distortions (i.e., deformations) of trigonal symmetry (symmetry group \( G = D_3 \)) or tetragonal symmetry (symmetry group \( G = D_4 \)) are introduced in order to describe a greater number of physical situations. Along these lines, he calculated the crystal-field matrices for the \( d^2 \) and \( d^3 \) configurations in tetragonal and trigonal symmetries.
From the group-theoretical point of view, the problem addressed by Schoenfeld and Rosengarten, and in a more complete way by Flato, concerns the labelling of the state vectors arising from the $d^N$ configuration via the use of a chain of groups starting from the classification group $U(5)$ and ending with the point symmetry group $G$ or its spinor group $G^*$ with $G \sim G^*/Z_2$. The head group $U(5)$ corresponds to the fact that the $2d + 1 = 5$ orbitals associated to the $d$–shell (for which $\ell = 2$) may undergo a unitary transformation without changing the spectrum of the relevant Hamiltonian. The end group is $G$ (the geometrical symmetry group for the ion in its environment) or $G^*$ (the spectral group that labels the spin, called the double group of $G$ in the terminology of H.A. Bethe) depending on the parity, even or odd, of the number of electrons $N$. Therefore, Flato used the chains

$$U(5) \supset SO(3) \supset O \supset D_3 \text{ or } D_4$$

and

$$U(5) \supset SU(2) \supset O^* \supset D_3^* \text{ or } D_4^*$$

for the configurations $d^2$ and $d^3$, respectively.

One of the advantages in using such chains of groups is that the Coulomb and spin-orbit matrices already calculated for the free ion can be easily transferred to the case of the ion in its surrounding. Indeed, the chain $U(5) \supset SO(3)$ or $U(5) \supset SU(2)$ is useful for the construction of the Coulomb and spin-orbit matrices. From a qualitative point of view, the chains $SO(3) \supset O \supset D_3 \text{ or } D_4$ and $SU(2) \supset O^* \supset D_3^* \text{ or } D_4^*$ serve to describe the (external) symmetry breaking (arising from an inhomogeneous Stark effect) when going from the free ion to the ion in its environment. From a quantitative point of view, the calculation of the crystal-field matrix can be done via the Wigner-Eckart theorem for the group $SU(2)$ in a nonstandard basis, namely a basis adapted to the chain $SO(3) \supset O \supset D_3 \text{ or } D_4$ for the $d^2$ configuration and the chain $SU(2) \supset O^* \supset D_3^* \text{ or } D_4^*$ for the $d^3$ configuration.

To fully understand the originality of the works by Flato, one has to realize that at this time most of the works on crystal- and ligand-field theories for $d^N$ ions in finite symmetries (involving both an idealized or high symmetry and a distortion or low symmetry) were conducted according to the approach by Tanabe, Sugano and Kamimura (from the Japanese school of M. Kotani) [7-10] and the one by Griffith [11-18]. In these approaches, the end group, i.e., the point symmetry group $G$ or its spinor group $G^*$, is considered as an isolated group so that the subduction (or descent in symmetry)

$$SO(3) \supset H \supset G \text{ or } SU(2) \supset H^* \supset G^*$$
is not fully taken into account \((H)\) stands for a high symmetry group, like \(O\), and \(G\) for a low symmetry group, like \(D_3\): the information about the descent in symmetry \(H \supset G\) or \(H^* \supset G^*\) is kept at the minimal level and the link with \(SO(3)\) or \(SU(2)\) is not completely exploited. Therefore, in the Griffith [11-18] and Tanabe, Sugano and Kamimura [7-10] approaches, all the necessary matrix elements must be calculated through the use of the Wigner-Racah algebra of \(G\) or \(G^*\). As a consequence, the calculation of the crystal-field energy matrix is very easy but the calculation of the Coulomb and spin-orbit energy matrices is very complicated. As a matter of fact, the invariance of the Coulomb and spin-orbit interactions under \(SO(3)\), when translated in terms of \(H \supset G\), leads to sums of invariant irreducible tensor operators which are difficult to handle via the Wigner-Eckart theorem for the group \(G\) or \(G^*\). This difficulty does not appear in the approach followed by Schoenfeld, Rosengarten and Flato.

The M.Sc. work by Flato [1] constituted the first systematic attempt to combine the simplifications afforded by the various chains of groups of the type \(U(5) \supset SO(3) \supset H \supset G\) and \(U(5) \supset SU(2) \supset H^* \supset G^*\) for dealing with partly-filled shell ions in discrete symmetry. The works by Schoenfeld, Rosengarten and Flato were used by Low and several of his collaborators for understanding optical spectra and electron paramagnetic resonance spectra of transition-metal ions in crystals (for example, see Refs. [6,19-21]). Although Moshé presented his M.Sc. thesis dissertation in 1960, several years later the subject was still up to date since he published a part of his thesis in 1965 [22].

The 1965 Flato paper and some impulse given by Moshé opened an avenue of new investigations in: (i) the Wigner-Racah algebra of the group \(SU(2)\) in bases adapted to chains of the type \(SU(2) \supset G^*\) where \(G^*\) is a group or a chain of groups of interest in molecular physics and condensed matter physics and (ii) the development of models in optical spectroscopy (mainly luminescence spectroscopy and photoemission spectroscopy) of \(d^N\) and \(f^N\) ions in crystalline fields (see Refs. [23-32] and references therein). Along this vein, in 1966 Moshé Flato suggested to a student, Maurice Kibler, from the ‘Faculté des Sciences de l’Université de Lyon’, to extend his M.Sc. thesis work to the situation where a magnetic interaction (arising from the Zeeman effect) is added to the Coulomb + spin-orbit + crystal-field Hamiltonian. This suggestion led the present author to develop the Wigner-Racah algebra of an arbitrary finite group embedded in \(SU(2)\) and to construct a phenomenological model, where symmetry considerations play a central role, for ions in a crystal subjected to an external magnetic field [23, 24, 27]. The research in that direction continued during several years with some students and collaborators of Kibler, especially
in:
- the study of specific chains of groups,
- the application of symmetry adaptation to electron paramagnetic resonance,
- the unification of crystal-field and ligand-field theories,
- the development of models for photo-electron spectroscopy and
- the derivation of a phenomenological approach to multi-photon electronic spectroscopy.

In 1995, with the advent of strong technological progress in two-photon absorption spectroscopy of rare earth ions doped materials, part of the above-mentioned works were revived in view of the elaboration of a model for analysing intensities of two-photon transitions between states of opposite parities for an $\ell^N$ ion in finite symmetry.

The rest of this paper is devoted to some personal recollections concerning the passage of Moshé in Lyon and to a brief description (in the appendix) of the basic ingredients for symmetry adaptation techniques in molecular and condensed matter spectroscopy developed by the present author more than thirty years ago under the direction of Moshé.

**Moshé in Lyon**

The arrival of Moshé in Lyon in 1964 has been a great event for many people. Although Moshé did not yet have a ‘Doctorat d’État’ (he submitted his ‘thèse de doctorat ès sciences physiques’ at ‘la Sorbonne’ in Paris on June 15, 1965), he joined the university staff of the ‘Université de Lyon’ as an associated professor in Physics under the recommendation of Jean Braconnier, ‘doyen de la Faculté des Sciences de Lyon’. (Jean Braconnier had been a member of the Bourbaki group.) He kept his position of associated professor for three years, sharing his life between Lyon and Paris. From 1964 to 1967, it was a real pleasure to enjoy the presence in Lyon of Moshé and Daniel (Daniel Sternheimer), almost every Monday and Tuesday to the best of my recollection. During the academic year 1964-65, Moshé gave master lectures on group theory both for finite groups and continuous groups. For most people in the audience the way to treat the subject was quite new (discussion at two levels: mathematical exposure and illustration from many branches of physics) and the lectures were very successful. The next two academic years, Moshé undertook to give a complete panorama of modern physics starting from classical mechanics up to quantum field theory (with intermediate steps in relativistic mechanics, quantum mechanics and statistical mechanics). The corresponding lectures were given by Moshé with an extraordinary economy principle in the presentation, a high level content and a constant preoccupation of the understanding by the audience.
During this short period (1964-67), several students (D. Allouch e, S. Ekong and J. Sternheimer) achieved their ‘thèse de doctorat de spécialité (troisième cycle)’ and two other students (J. Gréa and M. Kibler) started to work on their ‘thèse d’État’ under the supervision of Moshé.

Many of us in Lyon (students, collaborators and friends) have greatly benefited from Moshé not only for a transmission of a high level knowledge but also for having made possible to share with him a part of his dynamism, his enthusiasm, his love of physics (following Christian Frønsdal, one can add he was a physicist and a mathematician, teaching mathematics and having a passion for physics) and his respect of others. We also learned from Moshé how to be demanding for ourselves. His generosity (in all aspects of life) and his so particular way to consider each of us as a unique component of the world are qualities which should inspire us all.

Appendix : Finite Symmetry Adaptation

Let $G^*$ be a (finite) subgroup of $SU(2)$. We use $\Gamma (= \Gamma_0, \Gamma_1, \Gamma_2, \cdots)$ to denote an irreducible representation class (IRC) of $G^*$, $\Gamma_0$ being the identity IRC of $G^*$. We write $[\Gamma]$ for the dimension of a matrix representation associated to $\Gamma$. Such a matrix representation can be spanned by vectors of type

$$|ja\Gamma\gamma\rangle := \sum_{m=-j}^{j} |jm\rangle (jm|ja\Gamma\gamma\rangle) \tag{1}$$

which are linear combinations of vectors $|jm\rangle$ adapted to the chain $SU(2) \supset U(1)$. In Eq. (1), the branching label $a$ has to be used when the IRC $\Gamma$ of $G^*$ occurs several times in the IRC $(j)$ of $SU(2)$; this external multiplicity label may be described, at least partially, by IRC’s of subgroups of $SU(2)$ that contain in turn the group $G^*$. Furthermore, the label $\gamma$ is necessary when $[\Gamma] > 1$; it may be described sometimes by IRC’s of subgroups of $G^*$. Equation (1) is of a group theoretical nature: the reduction coefficients $(jm|ja\Gamma\gamma\rangle)$ depend on the chain $SU(2) \supset G^*$ but not on Physics. They may be chosen, thanks to Schur’s lemma, in such a way that the matrix representation associated to $\Gamma$ is the same for all $j$’s and $a$’s. Equation (1) defines the $\{ja\Gamma\gamma\}$ scheme for the chain $SU(2) \supset G^*$ that is more appropriate in molecular or condensed matter spectroscopy than the $\{jm\}$ scheme for the chain $SU(2) \supset U(1)$. For $j, a$ and $\Gamma$ fixed, the set $\{|ja\Gamma\gamma\rangle : \gamma = 1, \cdots, [\Gamma]\}$ is a $G^*$–irreducible tensorial set of vectors associated to $\Gamma$. Similarly, from the $SU(2) \supset U(1)$
spherical tensor operators $U_q^{(k)}$, we define the operators
\[
U_{a q}^{(k)} := \sum_{q=-k}^{k} U_{a q}^{(k)} (kq|ka\Gamma) \tag{2}
\]
so that, for $k$, $a$ and $\Gamma$ fixed, the set \( \{ U_{a q}^{(k)} : \gamma = 1, \cdots, [\Gamma] \} \) is a $G^*$-irreducible tensorial set of operators associated to $\Gamma$. The latter $G^*$-irreducible tensorial sets are also labelled by IRC’s of $SU(2)$ and, therefore, we can easily generate, by direct sum, nonstandard $SU(2)$-irreducible tensorial sets. Thus, we may apply the Wigner-Eckart theorem for the group $SU(2)$ in a nonstandard basis adapted to its subgroup $G^*$. As a result, we have
\[
(\tau_1 j_1 a_1 \Gamma_1 \gamma_1 | U_{a q}^{(k)} | \tau_2 j_2 a_2 \Gamma_2 \gamma_2) = (\tau_1 j_1 || U^{(k)} || \tau_2 j_2)
\]
\[
\times \sum_{a_3 \Gamma_3 \gamma_3} \begin{pmatrix} j_1 \\ a_3 \Gamma_3 \gamma_3 \\ a_1 \Gamma_1 \gamma_1 \end{pmatrix} \mathcal{J} \begin{pmatrix} j_1 k j_2 \\ a_3 \Gamma_3 \gamma_3 \Gamma a_\Gamma \gamma a_2 \Gamma_2 \gamma_2 \end{pmatrix} \tag{3}
\]
with
\[
\mathcal{J} \begin{pmatrix} j_1 j_2 j_3 \\ a_1 \Gamma_1 \gamma_1 a_2 \Gamma_2 \gamma_2 a_3 \Gamma_3 \gamma_3 \end{pmatrix} := \sum_{m_1 m_2 m_3} \begin{pmatrix} j_1 j_2 j_3 \\ m_1 m_2 m_3 \end{pmatrix} \times (j_1 m_1 | j_1 a_1 \Gamma_1 \gamma_1)^* (j_2 m_2 | j_2 a_2 \Gamma_2 \gamma_2)^* (j_3 m_3 | j_3 a_3 \Gamma_3 \gamma_3)^* \tag{4}
\]
and
\[
\begin{pmatrix} j \\ a_1 \Gamma_1 \gamma_1 a_2 \Gamma_2 \gamma_2 \end{pmatrix} := \sqrt{2j+1} \mathcal{J} \begin{pmatrix} j 0 j \\ a_1 \Gamma_1 \gamma_1 a_0 \Gamma_0 \gamma_0 a_2 \Gamma_2 \gamma_2 \end{pmatrix} \tag{5}
\]
On the right-hand side of (3), the quantum numbers $\tau_1$ and $\tau_2$, external to the chain $SU(2) \supset G^*$ (they depend on the physics of the considered problem), appear only in the reduced matrix element $(\tau_1 j_1 || U^{(k)} || \tau_2 j_2)$.

The $\mathcal{J}$ or 3-ja$\Gamma\gamma$ symbol in (4) is an $SU(2) \supset G^*$ symmetry adapted form of the usual 3-jm Wigner symbol. It constitutes a symmetrized form of the coefficient \[23\]
\[
f \begin{pmatrix} j_1 j_2 k \\ a_1 \Gamma_1 \gamma_1 a_2 \Gamma_2 \gamma_2 a_\Gamma \gamma \end{pmatrix} := \sum_{m_1 q m_2} (-1)^{j_1-m_1} \begin{pmatrix} j_1 k j_2 \\ -m_1 q m_2 \end{pmatrix} \times (j_1 m_1 | j_1 a_1 \Gamma_1 \gamma_1)^* (kq|ka\Gamma\gamma) (j_2 m_2 | j_2 a_2 \Gamma_2 \gamma_2) \tag{6}
\]
that generalizes the $f$ coefficient defined via
\[
(J\Gamma\gamma | U_{a_0}^{(k)} | J'\Gamma'\gamma') := \delta(\Gamma',\Gamma) \delta(\gamma',\gamma) f \begin{pmatrix} J J' k \\ \Gamma \Gamma' \Gamma_0 \end{pmatrix} \tag{7}
\]
in Moshé’s M.Sc. [1].
The 2-\(ja_1\Gamma_1\gamma_1\) symbol defined by (5) turns out to be an \(SU(2) \supset G^*\) symmetry adapted form of the Herring-Wigner metric tensor whose spherical components may be taken as
\[
\begin{pmatrix}
j \\
m_1 \\
m_2
\end{pmatrix} := (-1)^{j+m_1} \delta(m_2,-m_1).
\]
From equations (5) and (8) we have
\[
\begin{pmatrix}
j \\
a_1 \Gamma_1 \gamma_1 \\
a_2 \Gamma_2 \gamma_2
\end{pmatrix} = \sum_m (-1)^{j+m} (jm|ja_1 \Gamma_1 \gamma_1)^* (j,-m|ja_2 \Gamma_2 \gamma_2)^*.
\]
The metric tensor given by (9) allows us to handle all the phases occurring in the \(\{ja\Gamma\gamma\}\) scheme.

By combining (4), (6) and (9), we can rewrite (3) in the simple form
\[
(\tau_1 |ja_1 \Gamma_1 \gamma_1|U^{(k)}_{a_2 \gamma_2}|\tau_2) = (\tau_1 |ja_1 \Gamma_1 \gamma_1|U^{(k)}_{a_2 \gamma_2}|\tau_2) f
\begin{pmatrix}
j \\
a_1 \Gamma_1 \gamma_1 \\
a_2 \Gamma_2 \gamma_2 \\
a \Gamma \gamma
\end{pmatrix}.
\]
The interest of (3) and (10) for electronic spectroscopy of ions in crystalline fields has been discussed at length in Ref. [24]. From a mathematical viewpoint, it is to be observed that the factorization in (10) into the product of a \(G^*\)-dependent factor (the \(f\) coefficient) by a \(G^*\)-independent factor (the reduced matrix element) is valid whether the group \(G^*\) is multiplicity free or not. The internal multiplicity problem arising when \(G^*\) is not multiplicity free is thus easily solved by making use of (10).

An important property of the \(f\) coefficient is given by the following factorization formula which arises as a consequence of a lemma by Racah [5]. Indeed, we have
\[
f
\begin{pmatrix}
j_1 & j_2 & k \\
a_1 \Gamma_1 \gamma_1 & a_2 \Gamma_2 \gamma_2 & a \Gamma \gamma
\end{pmatrix} = (-1)^{2k} \frac{1}{\sqrt{2j_1 + 1}} \times \sum_\beta (j_2 a_2 \Gamma_2 + ka \Gamma_1 |j_1 a_1 \beta \Gamma_1)^* (\Gamma_2 \gamma_2 \Gamma_1 \beta \Gamma_1 |a j_1 a_1 \beta \Gamma_1)^*.
\]
where \((j_2 a_2 \Gamma_2 + ka \Gamma_1 |j_1 a_1 \beta \Gamma_1)\) is an isoscalar factor or reduced \(f\) symbol (independent of the labels \(\gamma_1, \gamma_2\) and \(\gamma\)) and \((\Gamma_2 \gamma_2 \Gamma_1 |a j_1 a_1 \beta \Gamma_1)^*\) a Clebsch-Gordan coefficient for the group \(G^*\) considered as an isolated entity. The label \(\beta\) is an internal multiplicity label to be used when the Kronecker product \(\Gamma_2 \otimes \Gamma\) is not multiplicity free.

Equations (10) and (11) are of central importance in the study of magnetic and electronic properties of an \(\ell^N\) ion in \(G\) (with \(G \sim G^*/Z_2\)) symmetry as well as in rotational spectroscopy of molecules invariant under \(G\). They proved to be useful in the development of phenomenological models for dealing with both energy levels and intensities of transitions. The reader may consult Refs. [23-26] for an application to crystal-field theory.
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