Group Theory With Applications in Chemical Physics

Group theory is widely used in many branches of physics and chemistry, and today it may be considered as an essential component in the training of both chemists and physicists. This book provides a thorough, self-contained introduction to the fundamentals of group theory and its applications in chemistry and molecular and solid state physics. The first half of the book, with the exception of a few marked sections, focuses on elementary topics. The second half (Chapters 11–18) deals with more advanced topics which often do not receive much attention in introductory texts. These include the rotation group, projective representations, space groups, and magnetic crystals. The book includes numerous examples, exercises, and problems, and it will appeal to advanced undergraduates and graduate students in the physical sciences. It is well suited to form the basis of a two-semester course in group theory or for private study.

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**To MFM
and to all those who love group theory**

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Preface

Symmetry pervades many forms of art and science, and group theory provides a systematic way of thinking about symmetry. The mathematical concept of a group was invented in 1823 by Évariste Galois. Its applications in physical science developed rapidly during the twentieth century, and today it is considered as an indispensable aid in many branches of physics and chemistry. This book provides a thorough introduction to the subject and could form the basis of two successive one-semester courses at the advanced undergraduate and graduate levels. Some features not usually found in an introductory text are detailed discussions of induced representations, the Dirac characters, the rotation group, projective representations, space groups, magnetic crystals, and spinor bases. New concepts or applications are illustrated by worked examples and there are a number of exercises. Answers to exercises are given at the end of each section. Problems appear at the end of each chapter, but solutions to problems are not included, as that would preclude their use as problem assignments. No previous knowledge of group theory is necessary, but it is assumed that readers will have an elementary knowledge of calculus and linear algebra and will have had a first course in quantum mechanics. An advanced knowledge of chemistry is not assumed; diagrams are given of all molecules that might be unfamiliar to a physicist.

The book falls naturally into two parts. Chapters 1–10 (with the exception of a few marked sections) are elementary and could form the basis of a one-semester advanced undergraduate course. This material has been used as the basis of such a course at the University of Western Ontario for many years and, though offered as a chemistry course, it was taken also by some physicists and applied mathematicians. Chapters 11–18 are at a necessarily higher level; this material is suited to a one-semester graduate course.

Throughout, explanations of new concepts and developments are detailed and, for the most part, complete. In a few instances complete proofs have been omitted and detailed references to other sources substituted. It has not been my intention to give a complete bibliography, but essential references to core work in group theory have been given. Other references supply the sources of experimental data and references where further development of a particular topic may be followed up.

I am considerably indebted to Professor Boris Zapol who not only drew all the diagrams but also read the entire manuscript and made many useful comments. I thank him also for his translation of the line from Alexander Pushkin quoted below. I am also indebted to my colleague Professor Alan Allnatt for his comments on Chapters 15 and 16 and for several fruitful discussions. I am indebted to Dr. Peter Neumann and Dr. Gabrielle Stoy of Oxford

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The Chemical Society of Japan, for Figure 10.3;
Taylor and Francis Ltd (http://www.tandf.co.uk/journals) for Table 10.2;
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The American Physical Society and Dr. C. J. Bradley for Table 14.6.

"Спуженье муз не терпит суеты … "   А. С. Пушкин
                                                            "19 октября"

which might be translated as:
"Who serves the muses should keep away from fuss," or, more prosaically,
"Life interferes with Art."

I am greatly indebted to my wife Mary Mullin for shielding me effectively from the daily intrusions of "Life" and thus enabling me to concentrate on this particular work of "Art."

Notation and conventions

General mathematical notation

$\equiv$	identically equal to
$\Rightarrow$	leads logically to; thus $p \Rightarrow q$ means if p is true, then q follows
$\sum$	sum of (no special notation is used when $\sum$ is applied to sets, since it will always be clear from the context when $\sum$ means a sum of sets)
$\forall$	all
iff	if and only if
$\exists$	there exists
$\overline{a}$	the negative of a (but note $\overline{\psi} = \Theta\psi$ in Chapter 13 and $\overline{R} = \overline{E}R$, an operator in the double group $\overline{G}$, in Chapter 8)
C^n	n-dimensional space in which the components of vectors are complex numbers
c, s	$\cos\phi$, $\sin\phi$
c_2, s_2	$\cos 2\phi$, $\sin 2\phi$
c x	$x\cos\phi$
c_n^m	$\cos(m\pi/n)$
i	imaginary unit, defined by $i^2 = \sqrt{-1}$
$q_1\, q_2\, q_3$	quaternion units
$\Re^n$	n-dimensional space, in which the components of vectors are real numbers
$\Re^3$	configuration space, that is the three-dimensional space of real vectors in which symmetry operations are represented
$s\, x$	$x\sin\phi$
$T(n)$	tensor of rank n in Section 15.1

Sets and groups

$\{g_i\}$	the set of objects $g_i, i = 1, \ldots, g$, which are generally referred to as 'elements'
$\in$	belongs to, as in $g_i \in G$
$\notin$	does not belong to
$A \to B$	map of set A onto set B
$a \to b$	map of element a (the pre-image of b) onto element b (the image of a)

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$A \cap B$	intersection of A and B, that is the set of all the elements that belong to both A and B
$A \cup B$	the union of A and B, that is the set of all the elements that belong to A, or to B, or to both A and B
G	a group $G = \{g_i\}$, the elements g_i of which have specific properties (Section 1.1)
E, or g_1	the identity element in G
g	the order of G, that is the number of elements in G
H, A, B	groups of order h, a, and b, respectively, often subgroups of G
$H \subset G$	H is a subset of G; if $\{h_i\}$ have the group properties, H is a subgroup of G of order h
$A \sim B$	the groups A and B are isomorphous
C	a cyclic group of order c
$\mathscr{C}_k$	the class of g_k in G (Section 1.2) of order c_k
C_{ij}^k	class constants in the expansion $\Omega_i \Omega_j = \sum\limits_{k=1}^{N_c} C_{ij}^k \Omega_{\overline{k}}$ (Section A2.2)
$g_i(\mathscr{C}_k)$	ith element of the kth class
$\underline{G}$	a group consisting of a unitary subgroup H and the coset AH, where A is an antiunitary operator (Section 13.2), such that $\underline{G} = \{H\} \oplus A\{H\}$
K	the kernel of G, of order k (Section 1.6)
l_i	dimension of ith irreducible representation
l_s	dimension of an irreducible spinor representation
l_v	dimension of an irreducible vector representation
N_c	number of classes in G
N_rc	number of regular classes
N_r	number of irreducible representations
N_s	number of irreducible spinor representations
N_v	number of irreducible vector representations
N(H|G)	the normalizer of H in G, of order n (Section 1.7)
t	index of a coset expansion of G on H, $G = \sum\limits_{r=1}^{t} g_r H$, with $g_r \notin H$ except for $g_1 = E$; $\{g_r\}$ is the set of coset representatives in the coset expansion of G, and $\{g_r\}$ is not used for G itself.
$Z(h_j\|G)$	the centralizer of h_j in G, of order z (Section 1.7)
Z_i	an abbreviation for $Z(g_i\|G)$
Ω_k, $\Omega(\mathscr{C}_k)$	Dirac character of $\mathscr{C}_k$, equal to $\sum\limits_{i=1}^{c_k} g_i(\mathscr{C}_k)$
$A \otimes B$	(outer) direct product of A and B, often abbreviated to DP
$A \boxtimes B$	inner direct product of A and B
$A \wedge B$	semidirect product of A and B
$A \overline{\otimes} B$	symmetric direct product of A and B (Section 5.3)
$A \underline{\otimes} B$	antisymmetric direct product of A and B (Section 5.3)

Vectors and matrices

$\mathbf{r}$	a polar vector (often just a vector) which changes sign under inversion; $\mathbf{r}$ may be represented by the directed line segment OP, where O is the origin of the coordinate system
$x\,y\,z$	coordinates of the point P and therefore the components of a vector $\mathbf{r} = $ OP; independent variables in the function $f(x, y, z)$.
$\mathbf{x}\,\mathbf{y}\,\mathbf{z}$	space-fixed right-handed orthonormal axes, collinear with OX, OY, OZ
$\mathbf{e}_1\,\mathbf{e}_2\,\mathbf{e}_3$	unit vectors, initially coincident with $\mathbf{x}\,\mathbf{y}\,\mathbf{z}$, but firmly embedded in configuration space (see R(ϕ $\mathbf{n}$) below). Note that $\{\mathbf{e}_1\,\mathbf{e}_2\,\mathbf{e}_3\}$ behave like polar vectors under rotation but are invariant under inversion and therefore they are *pseudovectors*. Since, in configuration space the vector $\mathbf{r} = \mathbf{e}_1 x + \mathbf{e}_2 y + \mathbf{e}_3 z$ changes sign on inversion, the components of $\mathbf{r}$, $\{x\,y\,z\}$, must change sign on inversion and are therefore *pseudoscalars*
$\{\mathbf{e}_i\}$	unit vectors in a space of n dimensions, $i = 1, \ldots, n$
$\{v_i\}$	components of the vector $\mathbf{v} = \sum_i \mathbf{e}_i v_i$
$\mathbb{A}$	the matrix $\mathbb{A} = [a_{rs}]$, with m rows and n columns so that $r = 1, \ldots, m$ and $s = 1, \ldots, n$. See Table A1.1 for definitions of some special matrices
$\mathbb{A}_{rs}$, a_{rs}	element of matrix $\mathbb{A}$ common to the rth row and sth column
$\mathbb{E}_n$	unit matrix of dimensions $n \times n$, in which all the elements are zero except those on the principal diagonal, which are all unity; often abbreviated to $\mathbb{E}$ when the dimensions of $\mathbb{E}$ may be understood from the context
det $\mathbb{A}$ or $|a_{rs}|$	determinant of the square matrix $\mathbb{A}$
$\mathbb{A} \otimes \mathbb{B}$	direct product of the matrices $\mathbb{A}$ and $\mathbb{B}$
$\mathbb{C}_{pr,qs}$	element $a_{pq} b_{rs}$ in $\mathbb{C} = \mathbb{A} \otimes \mathbb{B}$
$\mathbb{A}_{[ij]}$	ijth element (which is itself a matrix) of the supermatrix $\mathbb{A}$
$\langle a_1\ a_2 \ldots a_n|$	a matrix of one row containing the set of elements $\{a_i\}$
$\langle a|$	an abbreviation for $\langle a_1\ a_2 \ldots a_n|$. The set of elements $\{a_i\}$ may be basis vectors, for example $\langle \mathbf{e}_1\ \mathbf{e}_2\ \mathbf{e}_3|$, or basis functions $\langle \phi_1\ \phi_2 \ldots \phi_n|$.
$|b_1\ b_2 \ldots b_n\rangle$	a matrix of one column containing the set of elements $\{b_i\}$, often abbreviated to $|b\rangle$; $\langle b|$ is the transpose of $|b\rangle$
$\langle a'|$	the transform of $\langle a|$ under some stated operation
$\langle \mathbf{e}|r\rangle$	an abbreviation for the matrix representative of a vector $\mathbf{r}$; often given fully as $\langle \mathbf{e}_1\ \mathbf{e}_2\ \mathbf{e}_3 \mid x\,y\,z\rangle$

Brackets

$\langle\,|\,,\,|\,\rangle$	Dirac bra and ket, respectively; no special notation is used to distinguish the bra and ket from row and column matrices, since which objects are intended will always be clear from the context

$[A, B]$	commutator of A and B equal to $AB - BA$
$[a, A]$	complex number $a + \mathrm{i}A$
$[a\ ;\ \mathbf{A}]$	quaternion (Chapter 11)
$[g_i\ ;\ g_j]$	projective factor, or multiplier (Chapter 12); often abbreviated to $[i\ ;\ j]$
$[n_1\ n_2\ n_3]$	components of the unit vector $\mathbf{n}$, usually given without the normalization factor; for example, [1 1 1] are the components of the unit vector that makes equal angles with OX, OY, OZ, the normalization factor $3^{-\frac{1}{2}}$ being understood. Normalization factors will, however, be given explicitly when they enter into a calculation, as, for example, in calculations using quaternions

Angular momenta

$\mathbf{L}, \mathbf{S}, \mathbf{J}$	orbital, spin, and total angular momenta
$\hat{\mathbf{L}}, \hat{\mathbf{S}}, \hat{\mathbf{J}}$	quantum mechanical operators corresponding to $\mathbf{L}$, $\mathbf{S}$, and $\mathbf{J}$
L, S, J	quantum numbers that quantize $\mathbf{L}^2$, $\mathbf{S}^2$, and $\mathbf{J}^2$
$\hat{\mathbf{j}}$	operator that obeys the angular momentum commutation relations
$\mathbf{j} = \mathbf{j}_1 + \mathbf{j}_2$	total ($\mathbf{j}$) and individual ($\mathbf{j}_1, \mathbf{j}_2, \dots$) angular momenta, when angular momenta are coupled

Symmetry operators and their matrix representatives

A	antiunitary operator (Section 13.1); A, B may also denote linear, Hermitian operators according to context
E	identity operator
$\overline{E}$	operator $R(2\pi\ \mathbf{n})$ introduced in the formation of the double group $\overline{G} = \{R\ \overline{R}\}$ from G $= \{R\}$, where $\overline{R} = \overline{E}R$ (Section 8.1)
I	inversion operator
$I_1\ I_2\ I_3$	operators that generate infinitesimal rotations about $\mathbf{x}\ \mathbf{y}\ \mathbf{z}$, respectively (Chapter 11)
$\hat{I}_1\ \hat{I}_2\ \hat{I}_3$	function operators that correspond to $I_1\ I_2\ I_3$
$\mathtt{I}_3$	matrix representative of I_3, and similarly (note that the usual symbol $\Gamma(R)$ for the matrix representative of symmetry operator R is not used in this context, for brevity)
$\mathbf{I}$	generator of infinitesimal rotations about $\mathbf{n}$, with components I_1, I_2, I_3
$\mathtt{I}_{\mathbf{n}}$	matrix representative of $I_n = \mathbf{n} \cdot \mathbf{I}$
$\mathtt{J}_x\ \mathtt{J}_y\ \mathtt{J}_z$	matrix representatives of the angular momentum operators $\hat{J}_x,\ \hat{J}_y,\ \hat{J}_z$ for the basis $\langle m| = \langle \frac{1}{2},\ -\frac{1}{2}|$. Without the numerical factors of $\frac{1}{2}$, these are the Pauli matrices $\boldsymbol{\sigma}_1\ \boldsymbol{\sigma}_2\ \boldsymbol{\sigma}_3$

$R(\phi\ \mathbf{n})$	rotation through an angle ϕ about an axis which is the unit vector $\mathbf{n}$; here $\phi\ \mathbf{n}$ is not a product but a single symbol $\phi\mathbf{n}$ that fixes the three independent parameters necessary to describe a rotation (the three components of $\mathbf{n}$, $[n_1\ n_2\ n_3]$, being connected by the normalization condition); however, a space is inserted between ϕ and $\mathbf{n}$ in rotation operators for greater clarity, as in $R(2\pi/3\ \mathbf{n})$. The range of ϕ is $-\pi < \phi \leq \pi$. R acts on configuration space and on all vectors therein (including $\{\mathbf{e}_1\ \mathbf{e}_2\ \mathbf{e}_3\}$) (but not on $\{\mathbf{x}\ \mathbf{y}\ \mathbf{z}\}$, which define the space-fixed axes in the active representation)
$\hat{R}(\phi\ \mathbf{n})$	function operator that corresponds to the symmetry operator $R(\phi\ \mathbf{n})$, defined so that $\hat{R}f(\mathbf{r}) = f(R^{-1}\mathbf{r})$ (Section 3.5)
R, S, T	general symbols for point symmetry operators (point symmetry operators leave at least one point invariant)
$\hat{s}_x\ \hat{s}_y\ \hat{s}_z$	spin operators whose matrix representatives are the Pauli matrices and therefore equal to $\hat{J}_x,\ \hat{J}_y,\ \hat{J}_z$ without the common factor of $1/2$
T	translation operator (the distinction between T a translation operator and T when used as a point symmetry operator will always be clear from the context)
U	a unitary operator
$\mathscr{U}$	time-evolution operator (Section 13.1)
$\Gamma(R)$	matrix representative of the symmetry operator R; sometimes just R, for brevity
$\Gamma(R)_{pq}$	pqth element of the matrix representative of the symmetry operator R
Γ	matrix representation
$\Gamma_1 \approx \Gamma_2$	the matrix representations Γ_1 and Γ_2 are equivalent, that is related by a similarity transformation (Section 4.2)
$\Gamma = \sum_i c^i \Gamma_i$	the representation Γ is a direct sum of irreducible representations Γ_i, and each Γ_i occurs c^i times in the direct sum Γ; when specific representations (for example T_{1u}) are involved, this would be written $c(T_{1u})$
$\Gamma \supset \Gamma_i$	the reducible representation Γ includes Γ_i
$\Gamma_i = \sum_j c_{i,j}\ \Gamma_j$	the representation Γ_i is a direct sum of irreducible representations Γ_j and each Γ_j occurs $c_{i,j}$ times in the direct sum Γ_i
$\Gamma_{ij} = \sum_k c_{ij,k}\ \Gamma_k$	Clebsch–Gordan decomposition of the direct product $\Gamma_{ij} = \Gamma_i \boxtimes \Gamma_j$; $c_{ij,\ k}$ are the Clebsch–Gordan coefficients
$\sigma_\mathbf{n}$	reflection in the plane normal to $\mathbf{n}$
$\sigma_1\ \sigma_2\ \sigma_3$	the Pauli matrices (Section 11.6)
Θ	time-reversal operator

Bases

$\langle \mathbf{e}_1\ \mathbf{e}_2\ \mathbf{e}_3 \vert$	basis consisting of the three unit vectors $\{\mathbf{e}_1\ \mathbf{e}_2\ \mathbf{e}_3\}$ initially coincident with $\{\mathbf{x}\ \mathbf{y}\ \mathbf{z}\}$ but embedded in a unit sphere in configuration space so that $R\langle \mathbf{e}_1\ \mathbf{e}_2\ \mathbf{e}_3 \vert = \langle \mathbf{e}_1'\ \mathbf{e}_2'\ \mathbf{e}_3' \vert = \langle \mathbf{e}_1\ \mathbf{e}_2\ \mathbf{e}_3 \vert\ \Gamma(R)$. The 3×3 matrix $\Gamma(R)$ is the matrix representative of the symmetry operator R. Note that $\langle \mathbf{e}_1\ \mathbf{e}_2\ \mathbf{e}_3 \vert$ is often abbreviated to $\langle \mathbf{e} \vert$. If $\mathbf{r} \in \Re^3$, $R\ \mathbf{r} = R\langle \mathbf{e} \vert r\rangle = \langle \mathbf{e}' \vert r\rangle = \langle \mathbf{e} \vert \Gamma(R) \vert r\rangle = \langle \mathbf{e} \vert r'\rangle$, which shows that $\langle \mathbf{e} \vert$ and $\vert r\rangle$ are *dual bases*, that is they are transformed by the same matrix $\Gamma(R)$
$\langle R_x\ R_y\ R_z \vert$	basis comprising the three *infinitesimal* rotations R_x, R_y, R_z about OX, OY, OZ respectively (Section 4.6)
$\langle u^j_{-j} \ldots u^j_j \vert$	basis consisting of the $2j+1$ functions, u^j_m, $-j \le m \le j$, which are eigenfunctions of the z component of the angular momentum operator $\hat{J}_z$, and of $\hat{J}^2$, with the Condon and Shortley choice of phase. The angular momentum quantum numbers j and m may be either an integer or a half-integer. For integral j the u^j_m are the spherical harmonics $Y^m_l(\theta\ \varphi)$; $y^m_l(\theta\ \varphi)$ are the spherical harmonics written without normalization factors, for brevity
$\langle u^j_m \vert$	an abbreviation for $\langle u^j_{-j} \ldots u^j_j \vert$, also abbreviated to $\langle m \vert$
$\langle u\ \nu \vert$	spinor basis, an abbreviation for $\langle u^{\frac{1}{2}}_{\frac{1}{2}} \vert = \langle \vert\frac{1}{2}\ \frac{1}{2}\rangle\ \vert\frac{1}{2}\ -\frac{1}{2}\rangle \vert$, or $\langle \frac{1}{2}\ -\frac{1}{2} \vert$ in the $\langle m \vert$ notation
$\langle u'\ \nu' \vert$	transform of $\langle u\ \nu \vert$ in C^2, equal to $\langle u\ \nu \vert \mathbb{A}$
$\vert u\ \nu\rangle$	dual of $\langle u\ \nu \vert$, such that $\vert u'\ \nu'\rangle = \mathbb{A}\vert\ u\ \nu\rangle$
$\vert\ U_{-1}\ U_0\ U_1\rangle$	matrix representation of the spherical vector $\mathbf{U} \in C^3$ which is the dual of the basis $\langle y^{-1}_1\ y^0_1\ y^1_1 \vert$
N	normalization factor

Crystals

$\mathbf{a}_n = \langle \mathbf{a} \vert\ n\rangle$	lattice translation vector; $\mathbf{a}_n = \langle \mathbf{a}_1\ \mathbf{a}_2\ \mathbf{a}_3 \vert\ n_1\ n_2\ n_3\rangle$ (Section 16.1) (n is often used as an abbreviation for the $\mathbf{a}_n$)
$\mathbf{b}_m = \langle \mathbf{b} \vert\ m\rangle$	reciprocal lattice vector; $\mathbf{b}_m = \langle \mathbf{b}_1\ \mathbf{b}_2\ \mathbf{b}_3 \vert\ m_1\ m_2\ m_3\rangle = \langle \mathbf{e}_1\ \mathbf{e}_2\ \mathbf{e}_3 \vert\ m_x\ m_y\ m_z\rangle$ (Section 16.3); m is often used as an abbreviation for the components of $\mathbf{b}_m$

Abbreviations

1-D	one-dimensional (etc.)
AO	atomic orbital
BB	bilateral binary
bcc	body-centered cubic
CC	complex conjugate
CF	crystal field

CG	Clebsch–Gordan
CR	commutation relation
CS	Condon and Shortley
CT	charge transfer
DP	direct product
fcc	face-centered cubic
FE	free electron
FT	fundamental theorem
hcp	hexagonal close-packed
HSP	Hermitian scalar product
IR	irreducible representation
ITC	*International Tables for Crystallography* (Hahn (1983))
L, R	left and right, respectively, as in L and R cosets
LA	longitudinal acoustic
LCAO	linear combination of atomic orbitals
LI	linearly independent
LO	longitudinal optic
LS	left- side (of an equation)
LVS	linear vector space
MO	molecular orbital
MR	matrix representative
N	north, as in N pole
ORR	Onsager reciprocal relation
OT	orthogonality theorem
PBC	periodic boundary conditions
PF	projective factor
PR	projective representation
RS	right side (of an equation)
RS	Russell–Saunders, as in *RS* coupling or *RS* states
sc	simple cubic
SP	scalar product
TA	transverse acoustic
TO	transverse optic
ZOA	zero overlap approximation

Cross-references

The author (date) system is used to identify a book or article in the list of references, which precedes the index.

Equations in a different section to that in which they appear are referred to by eq. $(n_1 \cdot n_2 \cdot n_3)$, where n_1 is the chapter number, n_2 is the section number, and n_3 is the equation number within that section. Equations occurring within the same section are referred to simply by (n_3). Equations are numbered on the right, as usual, and, when appropriate,

a number (or numbers) on the left, in parentheses, indicates that these equations are used in the derivation of that equation so numbered. This convention means that such phrases as "it follows from" or "substituting eq. (n_4) in eq. (n_5)" can largely be dispensed with.

Examples and Exercises are referenced, for example, as Exercise $n_1 \cdot n_2\text{-}n_3$, even within the same section. Figures and Tables are numbered $n_1 \cdot n_3$ throughout each chapter. When a Table or Figure is referenced on the left side of an equation, their titles are abbreviated to T or F respectively, as in F16.1, for example.

Problems appear at the end of each chapter, and a particular problem may be referred to as Problem $n_1 \cdot n_3$, where n_1 is the number of the chapter in which Problem n_3 is to be found.