

Appendix A Effective Operator Calculations

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Effective operators are used in many types of calculations. They have found wide use in calculations relevant to the spectra of rare-earth systems, where calculations are carried out (as in Chapters 1 and 2) using matrix elements of operators between pure $4f^N$ states or between $4f^N$ and $4f^{N-1} 5d$ states.

The purpose of this appendix is to provide a brief introduction to the concept of effective operators and some of the issues that surround their use. A large literature exists and the interested reader is urged to refer to that literature for clarification of the many subtleties in this area (Brandow, 1967; Hurtubise and Freed, 1993, 1994; Bryson and Reid, 1998; Newman and Ng, 2000; Duan and Reid, 2001; Naguleswaran et al., 2003). In this Appendix we concentrate on explaining the concepts, rather than rigorous derivations.

A.1 Effective Hamiltonians and Effective Operators

Effective Hamiltonians and effective operators are defined within a model space M . In most cases of interest in this book the model space is the $4f^N$ configuration.

Consider eigenstates of the full Hamiltonian H ,

$$\mathcal{H} |a\rangle = E_a |a\rangle. \quad (\text{A.1})$$

The aim is to define an effective Hamiltonian H_{eff} that gives identical eigenvalues for model-space eigenstates $|a_0\rangle$, i. e.

$$\mathcal{H}_{\text{eff}} |a_0\rangle = E_a |a_0\rangle. \quad (\text{A.2})$$

Transformations may be defined that connect the full states and the model-space states:

$$|a\rangle = k |a_0\rangle \quad (\text{A.3})$$

and

$$|a_0\rangle = l |a\rangle. \quad (\text{A.4})$$

There is some flexibility (and many subtleties) in the choice of k and l . In the simplest case l is simply the projection operator for the model space (commonly written as P) and k is the wave operator (commonly written Ω). However, this choice can lead to non-intuitive behavior such as non-orthogonal model-space eigenstates and non-Hermitian effective operators. This is not a significant issue for most energy-level calculations, but is problematical for intensity calculations. Fortunately, it is possible to choose a canonical operator \hat{k} so that

$$|a\rangle = \hat{k} |a_0\rangle \quad (\text{A. 5})$$

and

$$|a_0\rangle = \hat{k}^\dagger |a\rangle. \quad (\text{A. 6})$$

Note that \hat{k} transforms between two spaces of different dimension, and while, $\hat{k}^\dagger \hat{k} = 1$, $\hat{k}^\dagger \hat{k} \neq 1$ (but is, instead, the model-space projection operator).

In our development we will use the \hat{k} operator. This has little effect at low order, but it should be noted that for higher orders of perturbation the requirement of Hermiticity and normalization leads to expressions that are more complex than those commonly found in quantum-mechanics texts.

In analogy to an effective Hamiltonian an effective operator (such as the operator for a dipole moment) has the same matrix elements between the model eigenstates as the full operator has between the full eigenstates, i. e.

$$\langle b_0 | A_{\text{eff}} | a_0 \rangle = \langle b | A | a \rangle. \quad (\text{A. 7})$$

This implies that the effective operator may be calculated from

$$A_{\text{eff}} = \hat{k}^\dagger A \hat{k}. \quad (\text{A. 8})$$

A.2 Perturbation Expansions

The transformations discussed in the previous section are seldom used directly. In a realistic calculation it is impossible to use the “full” space and the usual approach is to construct H_{eff} , \hat{k} , and A_{eff} using a time-independent Rayleigh-Schrödinger perturbation expansion. The Rayleigh-Schrödinger (rather than the Brillouin-Wigner) form is used so that the effective Hamiltonian and effective operators do not depend on the exact energies of the states. This has many important consequences, such as the symmetry arguments presented below. It is also essential to the development of a “linked-diagram” perturbation expansion (Hurtubise and Freed, 1994), the details of which we will not go into here. Perhaps the most obvious practical reason for using the Rayleigh-Schrödinger

expansion is that the energy-level and transition-intensity calculations discussed in Chapters 1 and 2 make use of matrices that need to be generated only once, then combined together, multiplied by phenomenological or *ab initio* values of the appropriate parameters. Dealing with matrices whose elements changed as the Hamiltonian parameters were varied would be completely impractical.

The first step in a perturbation-theory calculation is to partition the Hamiltonian into a zero-order part \mathcal{H}_0 and a perturbation V . The total Hamiltonian is

$$\mathcal{H} = \mathcal{H}_0 + V. \quad (\text{A. 9})$$

Eigenstates of \mathcal{H}_0 are labeled with Greek letters,

$$\mathcal{H}_0 | \alpha \rangle = E_\alpha^{(0)} | \alpha \rangle, \quad (\text{A. 10})$$

and the eigenstates of H with Roman letters,

$$\mathcal{H} | a \rangle = E_a | a \rangle. \quad (\text{A. 11})$$

An effective (e. g. crystal field) Hamiltonian H_{eff} may now be constructed to act within the model space M . The equations are simpler if all members of M are eigenstates of \mathcal{H}_0 with identical eigenvalues, E_0 , i. e. the model space is *degenerate*. This simplification will be used in what follows.

In the case of particular interest here the model space is the $4f^N$ configuration and a typical choice for H_0 is the Hartree-Fock Hamiltonian (see Chapter 1). The perturbation V is then a sum of several terms:

$$V = V_{\text{Coulomb}} + V_{\text{spin-orbit}} + V_{\text{CF}} + \dots, \quad (\text{A. 12})$$

This expression includes the non-spherical part of the potential arising from the Coulomb repulsion between the $4f$ electrons, the spin-orbit interaction, and the crystal field potential V_{CF} arising from the interaction of the ion with its ligands. The model-space eigenstates $| a_0 \rangle$ will be linear combinations of the $| 4f^N \alpha SLJM \rangle$ states

We require an effective Hamiltonian that gives identical eigenvalues for model-space states as the full Hamiltonian does for the complete states (Eqs. (A. 1) and (A. 2)). This leads to a Rayleigh-Schrödinger perturbation expansion for \mathcal{H}_{eff} (Hurtubise and Freed, 1994):

$$\begin{aligned} \mathcal{H}_{\text{eff}} = & \mathcal{H}_0 + V + \sum_{\beta \notin M} \frac{V | \beta \rangle \langle \beta | V}{E_0 - E_\beta^{(0)}} \\ & + \sum_{\gamma \notin M} \sum_{\beta \notin M} \frac{V | \gamma \rangle \langle \gamma | V | \beta \rangle \langle \beta | V}{(E_0 - E_\gamma^{(0)}) (E_0 - E_\beta^{(0)})} \end{aligned}$$

$$\begin{aligned}
 & -\frac{1}{2} \sum_{\beta \notin M} \sum_{\alpha \in M} \frac{V|\beta\rangle\langle\beta|V|\alpha\rangle\langle\alpha|V}{(E_0 - E_\beta^{(0)})^2} \\
 & -\frac{1}{2} \sum_{\alpha \in M} \sum_{\beta \notin M} \frac{V|\alpha\rangle\langle\alpha|V|\beta\rangle\langle\beta|V}{(E_0 - E_\beta^{(0)})^2} \\
 & + \dots
 \end{aligned} \tag{A.13}$$

where M refers to the model space. This expression is more symmetrical than expressions that the reader may encounter in quantum-mechanics textbooks. This symmetry is necessary to ensure that \mathcal{H}_{eff} is Hermitian.

The eigenstates of \mathcal{H}_{eff} (i. e. the phenomenological crystal-field eigenstates) are $|a_0\rangle$ and the exact states may be generated by

$$\begin{aligned}
 |a\rangle &= |a_0\rangle + \sum_{\beta \notin M} \frac{|\beta\rangle\langle\beta|V|a_0\rangle}{E_0 - E_\beta^{(0)}} \\
 &+ \sum_{\gamma \notin M} \sum_{\beta \notin M} \frac{|\gamma\rangle\langle\gamma|V|\beta\rangle\langle\beta|V|a_0\rangle}{(E_0 - E_\gamma^{(0)})(E_0 - E_\beta^{(0)})} \\
 &- \sum_{\beta \notin M} \sum_{\alpha \in M} \frac{|\beta\rangle\langle\beta|V|\alpha\rangle\langle\alpha|V|a_0\rangle}{(E_0 - E_\beta^{(0)})^2} \\
 &- \frac{1}{2} \sum_{\alpha \in M} \sum_{\beta \notin M} \frac{|\alpha\rangle\langle\alpha|V|\beta\rangle\langle\beta|V|a_0\rangle}{(E_0 - E_\beta^{(0)})^2} \\
 &+ \dots
 \end{aligned} \tag{A.14}$$

The last term is a normalization factor. In eqs. (A.13) and (A.14) we have carried the expansion to second order in V . Though in Chapters 1 and 2 we only make explicit use of the expressions up to first order in V it is important to realize that taking the calculations to higher order is a rather complex matter.

We can now use equation (A.14) to write the matrix elements of an effective operator (such as the dipole-moment operator), recalling that $E_i^{(0)} = E_f^{(0)} \equiv E_0$. To first order in V as the effective operator is

$$\begin{aligned}
 \langle b|A|a\rangle &= \langle b_0|A|a_0\rangle + \langle b_0|A \sum_{\beta \notin M} \frac{|\beta\rangle\langle\beta|V}{E_0 - E_\beta^{(0)}} |a_0\rangle \\
 &+ \langle b_0| \sum_{\beta \notin M} \frac{V|\beta\rangle\langle\beta|}{E_0 - E_\beta^{(0)}} A |a_0\rangle \\
 &+ \dots
 \end{aligned} \tag{A.15}$$

Higher-order expressions are easily derived from Eq. (A.14) (Hurtubise and Freed, 1994; Bryson and Reid, 1998).

We require an effective operator A_{eff} such that the matrix elements of A_{eff}

between eigenstates of H_{eff} are identical to the matrix elements of A between the exact eigenstates (if the expansion is carried to infinite order), i. e.

$$\langle b_0 | A_{\text{eff},g} | a_0 \rangle = \langle b | A | a \rangle. \quad (\text{A. 16})$$

The required operator may be read from Eq. (A. 15) as follows

$$A_{\text{eff}} = A + A \sum_{\beta \notin M} \frac{|\beta\rangle\langle\beta| V}{E_0 - E_\beta^{(0)}} + \sum_{\beta \notin M} \frac{V |\beta\rangle\langle\beta|}{E_0 - E_\beta^{(0)}} A + \dots \quad (\text{A. 17})$$

The denominators in the two summations in Eq. (A. 17) are equal term by term and if A is Hermitian then the effective operator is also Hermitian. We refer the reader to Hurtubise and Freed (1993, 1994) for a careful discussion showing that for Hermitian operators (such as the dipole moment operator) it is always possible to construct effective operators that are Hermitian Hurtubise and Freed (1993) and perturbation expansions that are Hermitian order by order in V Hurtubise and Freed (1994). Note that the right hand side of Eq. (A. 17) is only defined for the model space. We have omitted model-space projection operators from our equations for the sake of clarity.

A. 3 Symmetries and Selection Rules

An important feature of effective-Hamiltonian and effective-operator calculations is that if the canonical transformation \hat{k} is used then symmetries and selection rules for the full Hamiltonian also apply to the effective Hamiltonian (Hurtubise and Freed, 1993). If the Hamiltonian commutes with a symmetry operator S

$$[H, S] = 0 \quad (\text{A. 18})$$

then the model-space projections of H and S also commute:

$$[H_{\text{eff}}, S_{\text{eff}}] \equiv [\hat{k}^\dagger H \hat{k}, \hat{k}^\dagger S \hat{k}] = 0 \quad (\text{A. 19})$$

The existence of these commutation relations within the model space is essential for any application of symmetry arguments and is implicit in most treatments of crystal field calculations. Eq. (A. 19) implies that if the full Hamiltonian is symmetric under certain operations (rotations, reflections, Hermitian conjugation, time reversal, etc.), then so is the effective Hamiltonian.

It is important to realize that not all commutation relations apply in the model space. In general, commutators between operators that do not commute with H are not preserved under projection into the model space. An important example of this is the commutator of the position and momentum operators.

A.4 Implications

We have given a very brief introduction to the basic concepts of model spaces, effective Hamiltonians, and effective operators. If the canonical \hat{k} operator is used then in most cases model-space calculations work as expected. In particular, effective Hamiltonians are Hermitian and obey the symmetries of the full Hamiltonian. Hermitian operators give rise to Hermitian effective operators. Also, in a perturbation expansion using the Rayleigh-Schrödinger formalism, these properties are preserved order by order. The ability to construct effective Hamiltonians and effective operators that are Hermitian, and obey symmetries order by order in perturbation theory, is essential to applying symmetry arguments to determine the necessary effective operators. For example, in Chapter 1 we see that the crystal-field parameters B_q^k for $4f^N$ are only non-zero if k is even.

Many misconceptions permeate the literature surrounding the use of effective-operator (i. e. crystal-field) calculations in rare earth and actinide systems. We are now in a position to discuss some of these.

It is sometimes claimed that the crystal field model works for 4f-electron systems because the 4f orbitals do not mix strongly with the ligand states. This is not the case. An effective-Hamiltonian treatment will work even in the situation of large mixing with ligands (as in the case of the 5d orbitals of the $4f^{N-1}5d$ configuration). Consider, for example, an ion with one p electron and a single ligand on the Z axis. The p_z orbital may be strongly mixed with the ligand orbitals, whereas the p_x and p_y orbitals may be essentially pure p . From a molecular-orbital point of view this mixing will change the energy of the p_z -plus-ligand orbital relative to the other atomic orbitals. From a crystal-field point of view there would be a single parameter (B_0^2) governing this splitting. Both molecular-orbital and crystal-field pictures predict a splitting of the states. The magnitude of the mixing between the p_z and ligand orbitals can not be determined by merely measuring the energy-level splittings, but only from some other type of measurement, sensitive to the mixing, or from first-principles calculations.

Conversely, the fact that crystal-field fitting provides a satisfactory phenomenological model for rare earths and actinides is sometimes taken as evidence that the mixing with ligands is small, or that a simple point-charge calculation is adequate to explain the crystal field. This is also a flawed argument since the crystal-field model makes use only of symmetry to determine which parameters are non-zero. The sizes of the parameters must be compared to first-principles calculations if we want to know the size of various contributions point-charge, overlap, covalency, etc. (Ng and Newman, 1987; Newman and Ng, 2000) to the splittings.

For intensity calculations (Chapter 2) Eq. (A. 17) is used to calculate the dipole moment operator. Zero-order denominators, constant for each configuration, such as $4f^N$ or $4f^{N-1}5d$ appear in the calculation. This is often referred to as "closure". It is seen here to arise naturally from the perturbation expansion, rather than something that has to be imposed. Higher-order terms

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will, of course, modify the effective operators. These modifications could be interpreted as “correcting” the “closure assumption” of degenerate configurations. However, we prefer to think of them as simply higher-order terms in the perturbation expansion.

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Appendix B Matrix Elements of Tensor Operators

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Many of the calculations described in this book rely on the use of angular-momentum theory and group theory. There are a number of books available that give a complete introduction to angular-momentum calculations (Judd, 1963; Wybourne, 1965; Weissbluth, 1978; Lindgren and Morrison, 1986). The aim of this appendix is to give a brief introduction to allow the reader to make sense of the calculations described in Chapters 1 and 2.

B.1 Angular Momentum States and Operators

The essential concepts of angular momentum in quantum mechanics may be derived from considering basic angular momentum operators their eigenstates and their commutators. We begin with the general angular-momentum operators J_x , J_y and J_z , with well-known commutation relations

$$[J_x, J_y] = iJ_z, \quad (\text{B.1})$$

and cyclic permutations (we have set $\hbar = 1$). The operator $J^2 = J_x^2 + J_y^2 + J_z^2$ commutes with each of the, J_x , J_y , and J_z operators and it is traditional to use the simultaneous eigenstates of J^2 and J_z^2 as basis functions in atomic calculations. These basis states are labeled $|jm\rangle$ and have the following properties:

$$J_z |jm\rangle = m|jm\rangle, \quad (\text{B.2})$$

$$J^2 |jm\rangle = j(j+1)|jm\rangle, \quad (\text{B.3})$$

$$D(R)|jm\rangle = \sum_{m'} D_{m'm}^{(j)} |jm'\rangle \quad (\text{B.4})$$

where the last equation describes the behavior of the states under the rotation R , with $D(R)$ being the operator and $D_{m'm}^{(j)}$ the rotation matrix for a particular j .

B.2 Clebsh-Gordan Coefficients and 3-j Symbols

The coupling of angular momenta is a key concept that we require for any serious calculation. We may be coupling spin and orbital angular momentum, or the

angular momenta of two electrons. In either case, the idea is to create a linear combination of angular-momentum states that behave as an angular-momentum state, as defined by Eqs. (B. 2) – (B. 4). This is accomplished by the Clebsh-Gordan coefficients (sometimes called coupling coefficients) defined by

$$|jm\rangle = \sum_{m_1, m_2} |j_1 m_1\rangle |j_2 m_2\rangle \langle j_1 m_1, j_2 m_2 | jm\rangle. \quad (\text{B. 5})$$

It is often convenient to use the more symmetrical 3-j symbol, defined by

$$\begin{pmatrix} j_1 & j_2 & j_3 \\ m_1 & m_2 & m_3 \end{pmatrix} = (-1)^{j_1 - j_2 - m_3} \frac{1}{\sqrt{2j_3 + 1}} \langle j_1 m_1, j_2 m_2 | j_3 - m_3 \rangle. \quad (\text{B. 6})$$

In the derivation of selection rules (Chapters 1 and 2) the properties of the 3-j symbols play a key role. The 3-j symbol is zero unless $m_1 + m_2 + m_3 = 0$, $|m_1| < j_1$, $|m_2| < j_2$, $|m_3| < j_3$, and j_1, j_2 and j_3 form a triangle ($|j_1 - j_2| \leq j_3 \leq j_1 + j_2$).

B.3 Tensor Operators and the Wigner-Eckart Theorem

Tensor operators have angular-momentum quantum numbers, just like angular-momentum states. A tensor operator $T_q^{(k)}$ may be defined by its commutation relations, such as

$$[J_z, T_q^{(k)}] = q T_q^{(k)}, \quad (\text{B. 7})$$

or by its behavior under rotations

$$D(R) T_q^{(k)} D(R)^\dagger = \sum_{q'} D_{q'q}^{(k)} T_{q'}^{(k)}. \quad (\text{B. 8})$$

The latter equation emphasizes that the transformation properties of tensor operators and state kets are intimately related.

The matrix elements of tensor operators may be written in terms of the 3-j symbols as

$$\begin{aligned} & \langle \alpha JM | T_q^{(k)} | \alpha' J' M' \rangle T_q^{(k)} | \alpha' J' M' \rangle \\ & = (-1)^{J-M} \begin{pmatrix} J & k & J' \\ -M & q & M' \end{pmatrix} \langle \alpha J || T_q^{(k)} || \alpha' J' \rangle. \end{aligned} \quad (\text{B. 9})$$

The final bracket on the right hand side is known as a reduced matrix element. The key concept of the Wigner-Eckart theorem is that the “angular dependence” on the M quantum numbers is decoupled from the “physical” dependence of the

reduced matrix element.

The properties of the 3-j symbols discussed in the previous section give important selection rules on these matrix elements. For example, one-electron operators within the $4f^N$ configuration must have $k \leq 6$.

B.4 More Complex Situations

The calculations in Chapters 1 and 2 involve complicated angular momentum couplings of the $4f^N$ configuration, based on techniques pioneered by Racah over 50 years ago and discussed in detail in various books (Judd, 1963; Wybourne, 1965; Weissbluth, 1978; Lindgren and Morrison, 1986). We do not reproduce here all the equations involving the 3-j, 6-j and 9-j symbols, but emphasize that in principle the tools (and computer programs) exist to carry out such calculations. Thus we are free to concentrate on the physical meaning of the results.

As an example, we consider the one-electron $4f^N$ operators required for the crystalfield calculations of Chapter 1 and the intensity calculations of Chapter 2. The commonly-used states are labeled by the $\alpha SLJM$ quantum numbers, where α are extra labels required in complex configurations which may have several states with the same S and L . Reduced matrix elements of the unit tensor operators $U^{(k)}$ may be found in the tables of Nielson and Koster (1963) or in computer data files (see Appendix 3). These reduced matrix elements are at the SL level $\langle \alpha SL \| U^{(k)} \| \alpha' S' L' \rangle$. To obtain reduced matrix elements at the J level we must apply the Wigner-Eckart theorem in the following form (Judd, 1963; Weissbluth, 1978)

$$\langle \alpha SLJ \| U^{(k)} \| \alpha' S' L' J' \rangle = \delta(S, S') (-1)^{S'+L'+J+k} \sqrt{(2J+1)(2J'+1)} \\ \times \begin{Bmatrix} J & k & J' \\ L' & S & L \end{Bmatrix} \langle \alpha SL \| U^{(k)} \| \alpha' S' L' \rangle. \quad (\text{B. 10})$$

Eqs. (B.9) and (B.10) may be combined to obtain an expression for $\langle \alpha SLJM \| U_q^{(k)} \| \alpha' S' L' J' M' \rangle$.

The crystal-field parametrization used in Chapter 1 uses the spherical tensor operators $C_q^{(k)}$, rather than the unit tensor operators. However, the matrix elements are simply related and to the matrix elements of the $C_q^{(k)}$ within the $4f^N$ configuration are obtained from matrix elements of the $U_q^{(k)}$ by multiplying by

$$(-1)^l (2l+1) \begin{pmatrix} l & k & l \\ 0 & 0 & 0 \end{pmatrix}, \quad (\text{B. 11})$$

with $l=3$ for $4f$ electrons.

The spherical tensor operators differ from spherical harmonics by only a factor of $\sqrt{4\pi/(2k+1)}$. The lowest-rank spherical tensor operators may be

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written in cartesian form as

$$\begin{aligned}C_0^{(0)} &= 1 \\C_0^{(1)} &= z \\C_{\pm 1}^{(1)} &= \mp \sqrt{\frac{1}{2}}(x \pm iy) \\C_0^{(2)} &= \sqrt{\frac{1}{4}}(3z^2 - r^2) \\C_{\pm 1}^{(2)} &= \mp \sqrt{\frac{3}{2}}z(x \pm iy) \\C_{\pm 2}^{(2)} &= \sqrt{\frac{3}{8}}(x \pm iy)^2.\end{aligned}\tag{B. 12}$$

These expressions are often useful for working out selection rules and symmetry operations.

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