

Theory of neutron scattering by atomic electrons: jj-coupling scheme

This article has been downloaded from IOPscience. Please scroll down to see the full text article.

1991 J. Phys.: Condens. Matter 3 7095

(<http://iopscience.iop.org/0953-8984/3/37/001>)

View [the table of contents for this issue](#), or go to the [journal homepage](#) for more

Download details:

IP Address: 171.66.16.147

The article was downloaded on 11/05/2010 at 12:32

Please note that [terms and conditions apply](#).

Theory of neutron scattering by atomic electrons: *jj*-coupling scheme

Ewald Balcar† and Stephen W Lovesey‡§

† Atominstutute of the Austrian Universities, A-1020 Vienna, Austria

‡ Rutherford Appleton Laboratory, Oxfordshire OX11 0QX, UK

§ Institute of Physics, Uppsala University, S-751 21, Sweden

Abstract. Expressions are reported for the matrix element of the neutron–electron interaction for atomic electrons in a j^n configuration, appropriate for palladium and platinum group compounds and rare earth and actinide materials. For the latter, f -electron systems, an isolated ion is a realistic approximation. Compact expressions are provided, together with tables of reduced matrix elements, for elastic and inelastic structure factors, and compared with the corresponding Russell–Saunders expressions. Inelastic scattering by two f -electrons, including non-equivalent states, is presented in detail.

1. Introduction

Neutron scattering is a well proven technique for the study of magnetic properties of bulk matter; see for example Price and Sköld (1986). The scattering amplitudes for both elastic and inelastic events depend strongly on the magnitude of the scattering wavevector k : because the de Broglie wavelength matches the atomic dimensions. This feature is exploited to give information on the spatial properties and identify the atomic states engaged in scattering. Furthermore, neutron spectroscopy is not prescribed by the dipole selection rule. As a consequence, neutron atomic spectroscopy contains a wealth of information, some of which cannot be obtained using other techniques.

Bragg scattering is routinely used to provide accurate magnetization density maps which are confronted with theoretical calculations based on band theory or a local density approximation, e.g. Brown *et al* (1990) and Lindgren (1989). Spectroscopic studies of magnetic salts and metallic magnets directly provide crystal field energy levels (Stirling and McEwen 1987).

In the past few years the range of energies accessible for spectroscopy has increased significantly, so much so that transitions between intermultiplet atomic states (Coulomb excitations) can be studied. Thus far, experiments on rare earth and actinide (f^n) compounds and metals have been reported (Osborn *et al* 1991) and work on palladium ($4d^n$) and platinum ($5d^n$) groups of compounds might be expected. Observed transitions include both dipole allowed and dipole forbidden types; the latter are strongest (in f^n configurations) at intermediate wavevectors, typically $k \simeq 10 \text{ \AA}^{-1}$.

In the interpretation of diffraction and spectroscopic data for rare earth and actinide materials, cross sections for isolated ions are a meaningful reference point because $4f$ and $5f$ electron wavefunctions are relatively compact. Extensive calculations exist for electrons described by Russell–Saunders coupling (Balcar and Lovesey 1989,

Osborn *et al* 1991). Observed discrepancies between spectroscopic data and calculations prompt the question as to whether it might be more appropriate in some cases to describe the atomic states by *jj*-coupling. The *jj*-coupling is most likely to be realized in heavy atoms or in atoms which are multiply ionized. The reason for this is that the internal magnetic forces responsible for the one-electron spin-orbit couplings increase rapidly with the effective charge. Certain atoms may exhibit this type of coupling in excited states even though they do not in the normal level. No reliable calculations of the neutron cross section for electrons described by *jj*-coupling have been reported in the published literature. Work by Stassis and Deckman (1976) addresses the topic but their theory for inelastic events is known to be flawed (Balcar and Lovesey 1988).

Here we report a complete theory of the neutron amplitude for scattering from n equivalent atomic electrons described by *jj*-coupling. The results for elastic and inelastic structure factors are presented. General features are exposed and contrasted with corresponding results for Russell-Saunders coupling. The one-particle amplitudes we provide apply to j^1 and j^{2j} , and will also be required for the calculation of cross sections of states described by multi-component wavefunctions involving different j -states, such as those for atoms with d-electrons. Explicit values are tabulated for d- and f-electrons. Detailed results are given for the f^2 configuration, appropriate for praseodymium and uranium ions. This entails calculations for equivalent and non-equivalent electrons, i.e. transitions within a (j, j) term and between states which belong to (j, j) and (j, j') terms.

The neutron-electron interaction is taken to be of the standard form; no relativistic corrections are added as in the theory set out by Stassis and Deckman (1976). This stance is adopted on the grounds that with neutron-electron spectroscopy of Coulomb excitations still very much in its infancy more work, using the results of the present study, is required to gauge the possible importance of relativistic corrections in the scattering theory. The present work does permit the consistent use of radial wavefunctions evaluated from a Dirac-Fock scheme.

The neutron-electron interaction operator and cross sections are gathered in section 2. Key expressions for reduced matrix elements of electrons in the *jj*-coupling scheme are developed in section 3 and applied to the neutron scattering amplitude in section 4. Sections 5 and 6 report structure factors for j^n configurations for elastic and inelastic events, respectively. Applications of our results to f-electron systems, involving equivalent and non-equivalent states, are found in sections 7 and 8. Appropriate expressions for f^1 and f^{13} electrons are provided in section 9. A discussion of our findings for scattering by electrons described by a *jj*-coupling scheme is given in section 10.

2. Neutron-electron interaction and cross sections

The neutron-electron interaction is the sum of spin and orbital terms, which arise from the spin and current densities of unpaired electrons, respectively. Various derivations of the interaction are reviewed by Balcar and Lovesey (1989). For our present purpose it is convenient to introduce an operator

$$Q(\mathbf{k}) = \sum_j \exp(i\mathbf{k} \cdot \mathbf{R}_j) \left\{ \mathbf{s}_j - \frac{i}{\hbar k^2} (\mathbf{k} \times \mathbf{p}_j) \right\} \quad (2.1)$$

in which R_j , s_j , and p_j are the position, spin and momentum (conjugate to R_j) operators of the unpaired electron labelled by the index $j = 1, 2, \dots, n$. In the calculations reported here, these electrons are assumed to belong to a single atomic shell characterized by angular momentum l .

The initial and final atomic states are labelled by the composite quantum numbers μ, μ' . The partial differential neutron cross section for a scattering event in which the neutron energy changes by an amount $\hbar\omega = E - E'$ is

$$\frac{d^2\sigma}{d\Omega dE'} = \left(\frac{E'}{E}\right)^{1/2} r_0^2 \sum_{\alpha\beta} (\delta_{\alpha\beta} - k_\alpha k_\beta / k^2) \sum_{\mu\mu'} p_\mu \langle \mu | Q_\alpha^+ | \mu' \rangle \langle \mu' | Q_\beta | \mu \rangle \delta(\hbar\omega + E_\mu - E_{\mu'}). \quad (2.2)$$

Here α and β denote Cartesian components, $r_0 = -0.54 \times 10^{-12}$ cm is a standard unit for the scattering strength, and p_μ is the (normalized) probability for the initial state. In (2.2), and all subsequent examples, the neutron beam is assumed to be unpolarized, and we choose for the sake of brevity of notation to omit the explicit k -dependence of Q .

Bragg scattering is strictly elastic. The appropriate cross section is expressed in terms of the average scattering operator

$$\langle Q \rangle = \sum_{\mu} p_{\mu} \langle \mu | Q | \mu \rangle. \quad (2.3)$$

For a perfect crystal $\langle Q \rangle$ vanishes unless the neutron scattering vector k coincides with a reciprocal lattice vector, i.e. diffraction is observed when Bragg's law is satisfied. From (2.2) the diffraction cross section is readily found to be

$$\frac{d\sigma}{d\Omega} = r_0^2 \sum_{\alpha\beta} (\delta_{\alpha\beta} - k_\alpha k_\beta / k^2) \langle Q_\alpha^+ \rangle \langle Q_\beta \rangle. \quad (2.4)$$

In a simple collinear magnetic structure just one component of $\langle Q \rangle$ is finite, in which case the Bragg cross section vanishes when k is parallel to the preferred axis.

Another form of elastic scattering which is often of interest is that from a perfect paramagnet, realized to a good approximation at temperatures much in excess of the strength of coupling between magnetic ions. For many cases of interest $\langle Q_\alpha^+ Q_\beta \rangle$ vanishes unless $\alpha = \beta$, and $\langle |Q_\alpha|^2 \rangle$ is independent of the Cartesian index, so (2.2) reduces to

$$\frac{d\sigma}{d\Omega} = \frac{2}{3} r_0^2 \langle |Q|^2 \rangle. \quad (2.5)$$

In the limit of small scattering vectors it is well known that Q is proportional to the atomic moment. If the electron state is described by a total angular momentum J , we have

$$Q \rightarrow (g/2)J \quad k \rightarrow 0 \quad (2.6)$$

where g is the Landé factor. Using this limiting expression in (2.5), together with the result $J \cdot J = J(J+1)$, leads to the familiar cross section for the total scattering from an isolated paramagnetic ion,

$$\frac{d\sigma}{d\Omega} = (gr_0/2)^2 \frac{2}{3} J(J+1) \quad k \rightarrow 0. \quad (2.7)$$

The result (2.7) is valid for both Russell-Saunders and jj -coupling schemes but the Landé factors take different values, in the general case. For Russell-Saunders coupling g is expressed in terms of the total spin and orbital quantum numbers, S and L , whereas in jj -coupling

$$g = \frac{\{3j(j+1) + \frac{3}{4} - l(l+1)\}}{\{2j(j+1)\}}. \quad (2.8)$$

As an example, consider the ground state of praseodymium: f^2 ; $J = 4$, $L = 5$, $S = 1$ leads to $g = \frac{4}{5}$. In jj -coupling $j = \frac{5}{2}$ or $\frac{7}{2}$ in the first and second half of the f -electron series, respectively, and the corresponding Landé factors are $g = \frac{6}{7}$ and $\frac{8}{7}$.

As a final topic in this section we consider the cross section for inelastic scattering in which an atom is excited to a state at an energy Δ above the ground state. From (2.2)

$$\frac{d^2\sigma}{d\Omega dE'} = \left(\frac{E'}{E}\right)^{1/2} r_0^2 \mathcal{G}(J, J'; k) \delta(\hbar\omega - \Delta) \quad (2.9)$$

in which the structure factor is

$$\mathcal{G}(J, J'; k) = \sum_{\alpha\beta} (\delta_{\alpha\beta} - k_\alpha k_\beta / k^2) \sum_{\mu\mu'} p_\mu \langle \mu | Q_\alpha^+ | \mu' \rangle \langle \mu' | Q_\beta | \mu \rangle. \quad (2.10)$$

Very often a single crystal is not available. In this instance \mathcal{G} is averaged over all directions of k . The average total cross section for a particular transition is obtained from (2.9) using

$$\mathcal{G}(J, J'; k) = \int (d\hat{k}/4\pi) \sum_{\alpha\beta} (\delta_{\alpha\beta} - k_\alpha k_\beta / k^2) \sum_{\mu\mu'} p_\mu \langle \mu | Q_\alpha^+ | \mu' \rangle \langle \mu' | Q_\beta | \mu \rangle \quad (2.11)$$

in which the quantum numbers μ, μ' label the, possibly degenerate, states separated by an energy Δ .

In the following section we develop for electrons described by jj -coupling the matrix element $\langle \mu | Q | \mu' \rangle$ which occurs in the elastic and inelastic neutron cross sections we have reviewed.

3. Reduced matrix elements

We employ techniques pioneered by Racah which are today standard practice in atomic and nuclear spectroscopy; useful accounts are given by de-Shalit and Talmi (1963) and Judd (1963). Racah algebra is couched in terms of irreducible tensor operators. Hence, the first step is to express the interaction operator Q in terms of an appropriate tensor operator. Let us denote this operator by $X_{Q'}^{K'}$ where the positive integer K' is the rank of the tensor and $-K' \leq Q' \leq K'$. Explicit expressions for X are given in later sections. The matrix element of $X_{Q'}^{K'}$ has a structure dictated by the Wigner-Eckart theorem, namely

$$\langle JM | X_{Q'}^{K'} | J'M' \rangle = (-1)^{J-M} \begin{pmatrix} J & K' & J' \\ -M & Q' & M' \end{pmatrix} (J \| X^{K'} \| J') \quad (3.1)$$

where the right-hand side is, apart from a phase factor, the product of a $3j$ -symbol and a reduced matrix element. We use the phase conventions employed by Edmonds (1974) and also adopt his notation for the most part.

Attention is now shifted to the calculation of the reduced matrix element of X . For a configuration of n equivalent electrons, each with angular momentum j , fractional parentage coefficients are used to construct antisymmetric states. If $\chi(\alpha)$ denotes a one-electron operator, so that

$$X = \sum_{\alpha=1}^n \chi(\alpha) \quad (3.2)$$

then the reduced matrix element which occurs in equation (3.1) is

$$(\nu J \| X^{K'} \| \nu' J') = (j \| \chi^{K'} \| j) [K']^{-1/2} (\nu J \| V^{(K')} \| \nu' J'). \quad (3.3)$$

Here the first factor on the right-hand side is the reduced matrix element of χ with respect to a single-electron state and the explicit expression is provided in the latter parts of this section. The reduced matrix element of $V^{(K')}$ contains a sum of fractional parentage coefficients. Before giving the formula for the latter quantity note in (3.3) that ν denotes the seniority quantum number, and the convenient notation

$$[K, K', \dots] = (2K + 1)(2K' + 1) \dots$$

which is borrowed from Judd (1963).

In the definition of a fractional parentage coefficient $(\nu J \{ \bar{\nu} \bar{J} \})$ it is understood that $\bar{\nu} \bar{J}$ defines a parent state state of j^{n-1} whereas νJ is a state of j^n . These coefficients were tabulated by Bayman and Lande (1966) together with the decompositions of the configuration of j^n in terms of the states of the total angular momentum J . With the standard definition of a $6j$ -symbol it can be shown that

$$\begin{aligned} (\nu J \| V^{(K')} \| \nu' J') &= n [J, K', J']^{1/2} \sum_{\bar{\nu} \bar{J}} (\nu J \{ \bar{\nu} \bar{J} \}) (\nu' J' \{ \bar{\nu} \bar{J} \}) \\ &\times (-1)^{J+j+J+K'} \left\{ \begin{matrix} J & K' & J' \\ j & \bar{J} & j \end{matrix} \right\}. \end{aligned} \quad (3.4)$$

We now list three special cases for this reduced matrix element; some additional cases are covered by the results gathered in table 1. From the orthogonality relation for fractional parentage coefficients one obtains

$$(\nu J \| V^{(0)} \| \nu' J') = n [J]^{1/2} [j]^{-1/2} \delta_{JJ'} \delta_{\nu\nu'}. \quad (3.5)$$

The result

$$(\nu J \| V^{(1)} \| \nu' J') = \frac{\sqrt{3} (J \| J \| J)}{(j \| j \| j)} \delta_{JJ'} \delta_{\nu\nu'} \quad (3.6)$$

follows from (3.3) when X is taken to be the total angular momentum operator, in which case $\chi = j$. The last special case of immediate interest is that for $n = 2$. In

Table 1. Reduced matrix elements of $V^{(K')}$ defined in (3.4) for rare earth ions with $j = \frac{5}{2}$ and $\frac{7}{2}$. The squared values are given in prime factored form, where the asterisk indicates a negative value. An underlined digit represents a negative exponent for the corresponding prime: for example the entry $*3\bar{1}2\bar{1}$ is the number $-(2^3 \times 5^2/3 \times 7)^{1/2}$.

$Ce^{3+}/Sm^{3+} \quad n = 1$					
$\nu = 1 \quad J = \frac{5}{2}$	$J' = \frac{5}{2}$				
$(\nu J \ V^{(1)} \ \nu J')$	01				
$(\nu J \ V^{(3)} \ \nu J')$	0001				
$(\nu J \ V^{(5)} \ \nu J')$	0000,1				
$Pr^{3+}/Pm^{3+} \quad n = 2$					
$\nu = 2 \quad J = 4$	$J' = 4$	$J' = 2$			
$(\nu J \ V^{(1)} \ \nu J')$	320 $\bar{1}$				
$(\nu J \ V^{(3)} \ \nu J')$	*100 $\bar{1}$, 1	012 $\bar{1}$			
$(\nu J \ V^{(5)} \ \nu J')$	*000 $\bar{1}$, 11	*111 $\bar{1}$, 1			
$Nd^{3+} \quad n = 3$					
$\nu = 3 \quad J = \frac{9}{2}$	$J' = \frac{9}{2}$	$J' = \frac{3}{2}$			
$(\nu J \ V^{(1)} \ \nu J')$	020 $\bar{1}$, 1				
$(\nu J \ V^{(3)} \ \nu J')$	*010 $\bar{1}$, 11	*312 $\bar{1}$			
$(\nu J \ V^{(5)} \ \nu J')$	011 $\bar{1}$, 11	*211 $\bar{1}$, 1			
$Gd^{3+}/Yb^{3+} \quad n = 1$					
$\nu = 1 \quad J = \frac{7}{2}$	$J' = \frac{7}{2}$				
$(\nu J \ V^{(1)} \ \nu J')$	01				
$(\nu J \ V^{(3)} \ \nu J')$	0001				
$(\nu J \ V^{(5)} \ \nu J')$	0000, 1				
$(\nu J \ V^{(7)} \ \nu J')$	011				
$Tb^{3+}/Tm^{3+} \quad n = 2$					
$\nu = 2 \quad J = 6$	$J' = 6$	$J' = 2$	$J' = 4$		
$(\nu J \ V^{(1)} \ \nu J')$	0000, 01				
$(\nu J \ V^{(3)} \ \nu J')$	1101, 21		1002, 21		
$(\nu J \ V^{(5)} \ \nu J')$	*5100, 101	0111	0011, 1		
$(\nu J \ V^{(7)} \ \nu J')$	*0010, 2011	*101	*2210, 201		
$Dy^{3+}/Er^{3+} \quad n = 3$					
$\nu = 3 \quad J = \frac{15}{2}$	$J' = \frac{15}{2}$	$J' = \frac{11}{2}$	$J' = \frac{9}{2}$	$J' = \frac{5}{2}$	$J' = \frac{3}{2}$
$(\nu J \ V^{(1)} \ \nu J')$	1011, 001				
$(\nu J \ V^{(3)} \ \nu J')$	*1011, 1111	8001, 201	*7111, 21		
$(\nu J \ V^{(5)} \ \nu J')$	1001, 1211	*4011, 1111	*1111, 121	*1011, 31 (47) ²	
$(\nu J \ V^{(7)} \ \nu J')$	1010, 1211, 1	2110, 2111	*3511, 2211	3100, 311	*2001, 001
$Ho^{3+} \quad n = 4$					
$\nu = 4 \quad J = 8$	$J' = 8$	$J' = 5$	$J' = 4$	$J' = 2$	
$(\nu J \ V^{(1)} \ \nu J')$	2101, 001				
$(\nu J \ V^{(3)} \ \nu J')$	*1001, 1011	*2111, 101			
$(\nu J \ V^{(5)} \ \nu J')$	2001, 1211	0101, 0111	*2201, 021		
$(\nu J \ V^{(7)} \ \nu J')$	*0110, 1211, 1	2100, 1111	1201, 0211	*2111, 101	

this instance there is only one parent state $\bar{J} = j$, and the reduced matrix element of $V^{(K')}$ vanishes unless J and J' are even integers; $n = 2$,

$$(\nu J \| V^{(K')} \| \nu' J') = 2[J, K', J']^{1/2} \delta_{\nu\nu'} (-1)^{2j+K'} \left\{ \begin{matrix} J & K' & J' \\ j & j & j \end{matrix} \right\}. \quad (3.7)$$

This result is used in section 8 in the discussion of elastic and inelastic structure factors for f^2 .

Having summarized the apparatus for calculating the reduced matrix elements of a sum of one-electron operators for n equivalent electrons we proceed to apply it to the interaction operator defined in (2.1). From the presentational view-point it is convenient to split the work in two parts. First we present the reduced matrix elements of χ for the spin and orbital terms in the neutron-electron interaction and then, in the next section, we assemble the results to form the complete matrix element of Q for the configuration j^n . The one-electron reduced matrix elements presented in sections 3.1 and 3.2 can be used for problems that involve non-equivalent electrons, as demonstrated in section 8. Wavefunctions for ions in the palladium and platinum (d^n) groups of compounds usually contain several components, because the crystal field is influential. The cross sections for models of such systems can be calculated with the expressions reported in this and the following section.

3.1. Spin interaction

Using the standard expansion of a plane wave in terms of spherical harmonics and spherical Bessel functions

$$\begin{aligned} \exp(i\mathbf{k} \cdot \mathbf{R})s_q &= 4\pi \sum_{KQ} i^K j_K(kR) \{Y_Q^K(\hat{\mathbf{k}})\}^* s_q Y_Q^K(\hat{\mathbf{R}}) \\ &= (4\pi)^{1/2} \sum_{\substack{K,Q \\ K',Q'}} i^K j_K(kR) \{Y_Q^K(\hat{\mathbf{k}})\}^* (1qKQ|K'Q') \chi_{Q'}^{K'} \end{aligned} \quad (3.8)$$

where the second equality defines χ for the spin interaction, namely

$$\chi_{Q'}^{K'} = (4\pi)^{1/2} \sum_{qQ} s_q Y_Q^K(\hat{\mathbf{R}}) (1qKQ|K'Q') \quad (3.9)$$

in which $(1qKQ|K'Q')$ is a Clebsch-Gordan coefficient (related to a $3j$ -symbol, Edmonds 1974), $q = 0, \pm 1$ labels a spherical component of the electron spin, and $\hat{\mathbf{k}}$ and $\hat{\mathbf{R}}$ are unit vectors. From the definition (3.9) and use of standard formulae, the reduced matrix element is readily shown to be proportional to a $9j$ -symbol,

$$(j||\chi^{K'}||j') = (4\pi)^{1/2} [j, K', j']^{1/2} \begin{Bmatrix} \frac{1}{2} & l & j \\ \frac{1}{2} & l & j' \\ 1 & K & K' \end{Bmatrix} (\frac{1}{2}||s||\frac{1}{2})(l||Y^K||l). \quad (3.10)$$

In the case of n equivalent electrons $j = j'$. The more general expression (3.10) is required for non-equivalent electrons, discussed later in the paper, and the special case of one electron (hole).

3.2. Orbital interaction

The appropriate expression for the one-electron operator is

$$\chi_{Q'}^{K'} = (4\pi)^{1/2} \sum_{qQ} Y_Q^K(\hat{\mathbf{R}}) j_K(kR) (\hat{\mathbf{k}} \times \nabla)_q (KQ1q|K'Q'). \quad (3.11)$$

This operator is independent of the spin so its reduced matrix element is of the form

$$(j||\chi^{K'}||j') = (-1)^{j+\frac{1}{2}+l+K'} [j, j']^{1/2} \left\{ \begin{matrix} K' & j & j' \\ \frac{1}{2} & l & l \end{matrix} \right\} (l||\chi^{K'}||l). \quad (3.12)$$

At this juncture it is perhaps worthwhile to remind the reader that the formula for the reduced matrix element of an operator acting on two systems (e.g. spin and spatial) is arbitrary to within a phase factor. The latter depends on the chosen ordering of the individual operators, and the angular momentum coupling scheme. This phase factor must be consistent in (3.10) and (3.12). In our notation, the angular momentum coupling scheme is defined through a Clebsch-Gordan coefficient ($sm_s, lm_l | jm$), and the operators are written as in (3.10), namely spin to the left of orbital (spatial). The phase factor in (3.12) is consistent with this ordering, and use of a unit tensor for the spin component in (3.10).

The calculation of the reduced matrix element on the right-hand side of (3.12) is tedious; several approaches are discussed by Balcar and Lovesey (1989). We will simply lift the expression from this work, being careful to use exactly the same notation to facilitate the task for the reader who wishes to trace through the various steps. The result of interest is

$$(l||\chi^{K'}||l) = k\sqrt{6} [l]^2 (-1)^{(1+K+K')/2} [K, K']^{1/2} \\ \times A(K', K', l) \begin{pmatrix} 1 & K & K' \\ 0 & 0 & 0 \end{pmatrix} \left\{ \begin{matrix} 1 & 1 & 1 \\ K' & K & K' \end{matrix} \right\} [(j_{K'-1}) + (j_{K'+1})]. \quad (3.13)$$

In this expression,

$$(j_K) = \int_0^\infty r^2 dr f_j(r) j_K(kr) f_{j'}(r) \quad (3.14)$$

where $f_j(r)$ is the radial part of the one-electron wavefunction. The quantity $A(K', K', l)$ is defined by Balcar and Lovesey (1989). For now it is sufficient to note that K' is an odd integer, and the values of $A(K', K', l)$ required later are

$$A(1, 1, l) = \left\{ \frac{l(l+1)}{6(2l+1)^3} \right\}^{1/2} \quad (3.15)$$

and for $l = 3$,

$$A(3, 3, 3) = -\frac{1}{7^2} \sqrt{2} \quad A(5, 5, 3) = \frac{1}{11} \left\{ \frac{10}{7^3} \right\}^{1/2}. \quad (3.16)$$

The results (3.10), (3.12) and (3.13) could be obtained from equations (11.71) and (11.54) in Lovesey (1987), evaluated for a single electron.

The expressions (3.10), (3.12) and (3.13) together with (3.3) allow us to derive an explicit expression for the matrix element of Q in the configuration j^n . This topic is taken up in the next section. Results for non-equivalent electrons are presented in section 8.3.

4. $\langle j^n \nu JM | Q_q | j^n \nu' J' M' \rangle$

A compact way in which to present the matrix element of Q_q in the configuration j^n is to define a tensor operator $Z_{Q'}^{K'}$ on the lines of (3.8) and (3.11), and such that

$$Q_q = (4\pi)^{1/2} \sum_{\substack{K, Q \\ K', Q'}} (1qKQ | K'Q') \left\{ Y_Q^K(\vec{k}) \right\}^* Z_{Q'}^{K'}(K). \tag{4.1}$$

The matrix element of Q_q is therefore expressed in terms of a sum of the matrix elements of Z , and the latter satisfy the Wigner-Eckart theorem (3.1). The reduced matrix element of Z has a structure which is given by (3.3) in which the reduced matrix element of χ is, apart from some coefficients, the sum of (3.10) and (3.12) evaluated for $j = j'$.

Gathering the various expressions we find

$$\begin{aligned} (j^n \nu J || Z^{K'}(K) || j^n \nu' J') &= (j^n \nu J || V^{(K')} || j^n \nu' J') \\ &\times \left\{ \langle j_K \rangle \frac{E(K, K')}{(K' + 1)} + [\langle j_{K'-1} \rangle + \langle j_{K'+1} \rangle] \frac{F(K, K')}{(2K' + 1)} \right\} \end{aligned} \tag{4.2}$$

where $K = K' \pm 1$ and $K' = 1, 3, \dots, 2j$. The origin of the restrictions on K, K' are easily seen for the spin component $E(K, K')$. Turning to (3.10), the reduced matrix element of the spherical harmonic Y^K vanishes unless K is even, and the $9j$ -symbol, for $j = j'$, vanishes unless $1 + K + K'$ is even, and $1, K, K'$ satisfy the triangle condition. With regard to the orbital component $F(K, K')$, the $3j$ -symbol in (3.13) vanishes unless $1 + K + K'$ is even and $1, K, K'$ satisfy the triangle condition. The condition K' odd is contained in the definition of $A(K', K', l)$.

The reduced matrix element of $V^{(K')}$ for K' odd possesses some remarkable properties which are summarized in the identity,

$$(j^n \nu J || V^{(K')} || j^n \nu' J') = \delta_{\nu\nu'} (j^\nu \nu J || V^{(K')} || j^\nu \nu' J'). \tag{4.3}$$

This immediately reveals that the reduced matrix element is diagonal with respect to the seniority number and independent of the number of electrons n . Furthermore, it has the same sign and magnitude for the complementary states j^n and j^{2j+1-n} , which have common quantum labels including seniority number.

Using the results of the previous section,

$$E(K' + 1, K') = \left(\frac{K' + 1}{K'} \right)^{1/2} \beta(K') E(K' - 1, K') \tag{4.4}$$

in which

$$\beta(K') = \frac{-K'}{(K' + 1)} \times \begin{cases} \left(\frac{2j + 2 + K'}{2j + 1 - K'} \right) & j = l - \frac{1}{2} \\ \left(\frac{2j - K'}{2j + 1 + K'} \right) & j = l + \frac{1}{2} \end{cases} \tag{4.5}$$

and

$$F(K' + 1, K') = \left(\frac{K'}{K' + 1} \right)^{1/2} F(K' - 1, K'). \tag{4.6}$$

In view of these relations, (4.2) is completely determined by $E(K' - 1, K')$ and $F(K' - 1, K')$, which are quite simple expressions, namely,

$$E(K' - 1, K') = \frac{1}{2}(K' + 1) \left(\frac{K'}{2K' + 1} \right)^{1/2} \begin{pmatrix} l & l & K' - 1 \\ 0 & 0 & 0 \end{pmatrix} (-1)^{(2+2j+K')/2} \\ \times \begin{cases} [(2j + 1 - K')(2j + 2 - K')]^{1/2} & j = l - \frac{1}{2} \\ [(2j + K')(2j + 1 + K')]^{1/2} & j = l + \frac{1}{2} \end{cases} \quad (4.7)$$

and

$$F(K' - 1, K') = (2l + 1)(K' + 1)^{1/2} A(K', K', l) (-1)^{(K'-1)/2} \\ \times \begin{cases} [(2j + 2 + K')(2j + 1 - K')]^{1/2} & j = l - \frac{1}{2} \\ [(2j + 1 + K')(2j - K')]^{1/2} & j = l + \frac{1}{2} \end{cases} \quad (4.8)$$

where values for $A(K', K', l)$ appear in (3.15) and (3.16). In both these expressions the maximum $K' = 2j$. Values of $E(K, K')$ and $F(K, K')$ required for the description of scattering by electrons in a configuration f^n are listed in table 2.

Table 2. The quantities $E(K, K')$ and $F(K, K')$ defined in (4.7) and (4.8) for rare earth ions with $j = \frac{5}{2}$ and $j = \frac{7}{2}$. See table 1 for details.

$j = \frac{5}{2}$		$j = \frac{5}{2}$		$j = \frac{5}{2}$		$j = \frac{7}{2}$	
$E(0, 1)$	*1011	$E(0, 1)$	3101	$F(0, 1)$	5011	$F(0, 1)$	3301
$E(2, 1)$	6011	$E(2, 1)$	*4101	$F(2, 1)$	4011	$F(2, 1)$	2301
$E(2, 3)$	*6112	$E(2, 3)$	5002, 1	$F(2, 3)$	4112	$F(2, 3)$	5002, 1
$E(4, 3)$	6012	$E(4, 3)$	*7102, 1	$F(4, 3)$	2212	$F(4, 3)$	3102, 1
$E(4, 5)$	*2211, 2	$E(4, 5)$	3311, 21	$F(4, 5)$	4211, 2	$F(4, 5)$	3111, 21
$E(6, 5)$	5321, 2	$E(6, 5)$	*4221, 21	$F(6, 5)$	3121, 2	$F(6, 5)$	2021, 21
—	—	$E(6, 7)$	7121, 11	—	—	—	—
—	—	$E(8, 7)$	0	—	—	—	—

The results reported in this section so far amount to a complete prescription for the matrix element of the neutron-electron interaction (2.1) for the configuration j^n . The remainder of the section is taken up with discussion of some particular features of the main result.

Let us compare the reduced matrix element of Z with the corresponding expressions for Russell-Saunders coupling. Referring to our previous work on this subject (Lovesey 1987, Balcar and Lovesey 1989) the analogue of (4.2) may be written in the form

$$(f^n SLJ \| Z^{K'}(K) \| f^n S' L' J') = (-1)^{J' - J + K' + K} \left\{ \frac{3(2J + 1)}{(2K' + 1)} \right\}^{1/2} \\ \times \left\{ i^K \left(\frac{2K' + 1}{3} \right)^{1/2} C(K, K') + A(K, K') \right\}.$$

Here the spin reduced matrix element

$$C(K, K') \propto (SLJ \| W^{(1,K)K'} \| S' L' J')$$

and the orbital reduced matrix element

$$A(K, K') \propto (SLJ||W^{(0,K')K'}||S'L'J')$$

where $W^{(a,b)c}$ is the standard unit tensor operator; following the convention described in section 3, the indices a, b refer to the spin and orbital systems, respectively. For the extreme case of one electron the jj -coupling and Russell-Saunders schemes are identical given the identification of quantum labels $S = S' = \frac{1}{2}$, $L = L' = l$, and $J = j$, $J' = j'$. Hence, in this instance the one-electron reduced matrix elements $E(K, K')$ and $F(K, K')$ in (4.2) can be matched with $C(K, K')$ and $A(K, K')$; this topic is taken up again in section 9.

On approaching the forward direction of scattering $k \rightarrow 0$ the only significant radial integral (3.14) in the matrix element of Q_q is $\langle j_0 \rangle$, and this ultimately achieves the value unity while $\langle j_K \rangle$ for $K \neq 0$ vanish. By examining (4.2) in this limit we conclude that the matrix element for $k \rightarrow 0$ is proportional to $(\nu J||V^{(1)}||\nu J')$, and the result (3.6) shows that only the diagonal element is finite. Hence, for a configuration j^n ($n \geq 2$), inelastic events do not contribute to the scattering in the forward direction. This result contrasts with the situation for Russell-Saunders coupling, where elastic and dipole allowed ($J \rightarrow J \pm 1$) events contribute in the limit $k \rightarrow 0$. For $n = 1$ the calculations are based on the one-electron matrix elements provided in section 3.

The quantities $E(0, 1)$ and $F(0, 1)$ together with the reduced matrix element of $V^{(1)}$ determine the value of the forward scattering amplitude. A straightforward calculation based on (4.7) and (4.8) reveals that

$$E(0, 1) = 2 \frac{(J||S||J)}{(J||V^{(1)}||J)} = \frac{2}{\sqrt{3}}(j||s||j)$$

and

$$F(0, 1) = \frac{(J||L||J)}{(J||V^{(1)}||J)} = \frac{1}{\sqrt{3}}(j||l||j). \tag{4.9}$$

These two results are required to verify that the scattering in the forward direction is proportional to the square of the magnetic moment.

In many applications the cross section of interest is the total value for states J, J' averaged over all directions of the scattering vector k . The latter operation is certainly appropriate when a polycrystalline sample is used in an experiment, and summing over M, M' is a good approximation when the energy resolution available does not allow the fine structure of the states J, M and J', M' to be resolved. It can be shown that the quantity denoted by \mathcal{G} in (2.11) is

$$\begin{aligned} \mathcal{G}(J, J'; k) &= \int (d\hat{k}/4\pi) \sum_{MM'} (2J + 1)^{-1} \sum_{\alpha\beta} (\delta_{\alpha\beta} - k_\alpha k_\beta / k^2) \\ &\quad \times \langle \nu' J' M' | Q_\alpha | \nu J M \rangle^* \langle \nu' J' M' | Q_\beta | \nu J M \rangle \\ &= (2J + 1)^{-1} \sum_{K'} \frac{(\nu J || V^{(K')} || \nu' J')^2}{(K' + 1)(2K' + 1)} \{ E(K' - 1, K') (\langle j_{K'-1} \rangle \\ &\quad + \beta(K') \langle j_{K'+1} \rangle) + F(K' - 1, K') (\langle j_{K'-1} \rangle + \langle j_{K'+1} \rangle) \}^2 \end{aligned} \tag{4.10}$$

where $K' = 1, 3, \dots, 2j$. The factor $(1/(2J + 1))$ accounts for the degeneracy[†] of the initial state, i.e. it is the value of p_μ in the general formula (2.2).

The relative simplicity of (4.10) indicates that the expression for Q can be simplified to some extent. To this end note that the component of Q parallel to the wavevector k does not contribute to the cross section. Hence, any function proportional to k can be added to Q without changing the cross section. This non-uniqueness of Q can be exploited to remove the term $K = K' + 1$, say. Making use of (4.4) and (4.6) we obtain an alternative expression

$$\begin{aligned} \langle j^n \nu J M | Q_q | j^n \nu' J' M' \rangle &= (4\pi)^{1/2} \sum_{K' Q' Q} (-1)^{J-M} \begin{pmatrix} J & K' & J' \\ -M & Q' & M' \end{pmatrix} \\ &\times (1qK' - 1Q|K'Q') \left\{ Y_Q^{K'-1}(\vec{k}) \right\}^* (\nu J \| V^{(K')} \| \nu' J') \\ &\times \frac{1}{(K' + 1)} \left\{ E(K' - 1, K') (\langle j_{K'-1} \rangle + \beta(K') \langle j_{K'+1} \rangle) \right. \\ &\left. + F(K' - 1, K') (\langle j_{K'-1} \rangle + \langle j_{K'+1} \rangle) \right\}. \end{aligned} \quad (4.11)$$

The similarity of structure in expressions (4.10) and (4.11) should be noted. In both cases there are no terms with $\nu \neq \nu'$ because the reduced matrix element of $V^{(K')}$ for K' odd is diagonal in the seniority number.

For d^n configurations crystal field effects, in general, lead to multi-component wavefunctions. In consequence, the scattering amplitude contains several terms, each of which is evaluated with (4.11). On the other hand, for f^n configurations an isolated ion is frequently a quite realistic model. Not surprisingly, perhaps, calculations in this instance can be pushed much further along, in fact to the extent of obtaining compact expressions for the elastic and inelastic structure factors.

5. Elastic scattering: j^n

The structure factor for scattering from an isolated paramagnetic ion is given by (4.10) evaluated for $J = J'$. It is usual to cast the elastic structure factor in terms of an atomic form factor $\mathcal{F}(k)$ defined by

$$\mathcal{G}(J, J; k) = \mathcal{G}(J, J; 0) \mathcal{F}^2(k) \quad (5.1)$$

in which

$$\mathcal{G}(J, J; 0) = \frac{1}{6} g^2 J(J + 1)$$

follows from (2.8) and (4.9), cf (2.7). The result (5.1) is exact whereas by using (4.11) it follows that (2.5) is correct for terms involving $\langle j_0 \rangle$, i.e. at small wavevectors.

Experience with atoms which are well described by the Russell-Saunders scheme has shown that the so-called dipole approximation to the form factor furnishes a tolerable description of data. Referring to (4.10), this approximation would be obtained by neglecting terms beyond $K' = 1$ and the term proportional to $\beta(1)$. However, contrary to the Russell-Saunders case, the factor $\beta(1)$ is not small; for $j = \frac{5}{2}$ and $\frac{7}{2}$ the values of $\beta(1)$ are $(-\frac{4}{5})$ and $(-\frac{1}{3})$, respectively. In view of this, the following

approximation for $\mathcal{F}(k)$ is obtained from (5.1) using the first contribution in (4.10) including the term in $\beta(1)$. A straightforward calculation brings us to the result

$$\mathcal{F}(k) = \langle j_0 \rangle + \langle j_2 \rangle \times \begin{cases} (2j+3)/2j & j = l - \frac{1}{2} \\ (2j-1)/2(j+1) & j = l + \frac{1}{2} \end{cases} \quad (5.2)$$

The coefficient of $\langle j_2 \rangle$ is $\frac{8}{5}(\frac{2}{3})$ for $j = \frac{5}{2}(\frac{7}{2})$, and so this term appears to be most significant in the first half of the f-electron series. However, radial integrals depend on j so the j -dependence of the form factor is not a simple issue.

In an ordered magnetic material the ions are described by the state $M = J$ to a good approximation, i.e. the ions are close to magnetic saturation. Setting $M = M' = J' = J$ in (4.11),

$$\begin{aligned} \langle j^n \nu J J | Q_0 | j^n \nu J J \rangle &= (4\pi)^{1/2} \sum_{K'} \begin{pmatrix} J & K' & J \\ -J & 0 & J \end{pmatrix} (10K' - 10 | K' 0) \\ &\times (\nu J || V^{(K')} || \nu J) \frac{1}{K'+1} \{ E(K'-1, K') (\langle j_{K'-1} \rangle + \beta(K') \langle j_{K'+1} \rangle) \\ &+ F(K'-1, K') (\langle j_{K'-1} \rangle + \langle j_{K'+1} \rangle) \} Y_Q^{K'-1}(\hat{k}) \\ &= (4\pi)^{1/2} \sum_{K'} H(K') Y_0^{K'-1}(\hat{k}) = L(\hat{k}) \end{aligned} \quad (5.3)$$

where the second and third equalities define $H(K')$ and $L(\hat{k})$ which are convenient in subsequent work. Using spherical coordinates θ, ϕ for the unit vector \hat{k} , and introducing the quantity

$$M(\theta) = (4\pi)^{1/2} \sum_{K'=3} \left(\frac{K'-1}{K'} \right)^{1/2} H(K') e^{-i\phi} Y_1^{K'-1}(\theta, \phi) \quad (5.4)$$

the matrix elements of Q_x and Q_y are readily shown to be $M(\theta) \cos \phi$ and $M(\theta) \sin \phi$, respectively. Hence,

$$\begin{aligned} \sum_{\alpha\beta} (\delta_{\alpha\beta} - \hat{k}_\alpha \hat{k}_\beta) \langle J | Q_\alpha | J \rangle^* \langle J | Q_\beta | J \rangle \\ = \{ \sin \theta L(\theta) - \cos \theta M(\theta) \}^2 \rightarrow \left\{ \frac{1}{2} g J \sin \theta \right\}^2 \quad \text{for } k \rightarrow 0 \end{aligned} \quad (5.5)$$

where in the last line g is the Landé factor (2.8). The atomic form factor for a saturated ion, normalized to unity at $k = 0$ is

$$\mathcal{F}(\hat{k}) = (2/gJ) \{ L(\theta) - M(\theta) \cot \theta \} \quad (5.6)$$

which is independent of the angle ϕ , i.e. the form factor has cylindrical symmetry.

By making use of the identity

$$\begin{aligned} \{ \sin \theta L(\theta) - \cos \theta M(\theta) \} \\ = - (4\pi)^{1/2} \sum_{K'} \left\{ \frac{(K'+1)(2K'-1)}{K'(2K'+1)} \right\}^{1/2} H(K') e^{-i\phi} Y_1^{K'}(\theta, \phi) \end{aligned} \quad (5.7)$$

the average of the structure factor (5.5) with respect to the directions of \hat{k} , cf (2.11) and (4.10), is seen to be

$$\sum_{K'} \left\{ \frac{(K'+1)(2K'-1)}{K'(2K'+1)} \right\} H^2(K') = \frac{1}{6} \{gJ\mathcal{F}(\hat{k})\}^2. \quad (5.8)$$

Here $\mathcal{F}(\hat{k})$ is the form factor for the averaged structure factor of a saturated ion. This is not the same as the paramagnetic form factor defined in (5.1); a little algebra shows that $\mathcal{G}(J, J; \hat{k})$ derived from (4.10) can be written

$$\mathcal{G}(J, J; \hat{k}) = \sum_{K'} \left\{ \frac{(K'+1)(2K'-1)}{K'(2K'+1)} \right\} [H(K')/(K'0JJ|JJ)]^2 \quad (5.9)$$

which compared with (5.8) makes explicit the difference between paramagnetic and saturated ion form factors. However, they are exactly the same if terms beyond $K' = 1$ are neglected in (5.8) and (5.9), i.e. the dipole approximation (5.2) applies to paramagnetic and saturated magnetic ions.

If a single crystal (domain) is studied then (5.5) is the correct basis for interpretation in terms of scattering from an isolated saturated magnetic ion. In one standard scattering geometry, the preferred axis is perpendicular to the plane defined by the incident and scattered neutron wavevectors, in which case $\theta = \pi/2$ and the term M in (5.5), that arises from the x and y components of the interaction, is not observed.

The identity (5.7) provides a convenient route for the calculation of $\mathcal{F}(\hat{k})$, particularly when it is written in terms of Legendre polynomials,

$$\begin{aligned} \mathcal{F}(\hat{k}) &= (2/gJ) \sum_{K'} \frac{(2K'-1)^{1/2}}{K'} H(K') \frac{d}{dx} P_{K'}(x) \\ &= (2/gJ) \left\{ H(1) + \frac{\sqrt{5}}{2}(5x^2-1)H(3) + \frac{9}{8}(21x^4-14x^2+1)H(5) \right\}. \end{aligned} \quad (5.10)$$

Here, $x = \cos \theta$ and the last expression is correct for $j = \frac{5}{2}$. For $j \geq \frac{7}{2}$, it should be borne in mind that, although additional terms are added to the expression (5.10), the latter contains all contributions in the radial integrals $\langle j_0 \rangle$, $\langle j_2 \rangle$ and $\langle j_4 \rangle$, and hence it is likely to be more than adequate for most purposes.

6. Inelastic scattering: j^n

The cross section for a scattering event in which an isolated ion is promoted from a state JM to $J'M'$, at an energy Δ above the initial state, is calculated from (2.9) using (4.2) or (4.11). If the fine structure in the initial and final states, due to the total or partial lifting of the degeneracies in M or M' , is on an energy scale which is very small compared with Δ then it might be appropriate to average over M and sum over M' . Referring to expression (2.10) for the inelastic structure factors $\mu = M$, $\mu' = M'$ and $p_\mu = 1/(2J+1)$. In the event that the sample is a polycrystal and the structure factor is averaged over the directions of the scattering vector \hat{k} , as in (2.11), which leads to the result (4.10), for $\mathcal{G}(J, J'; \hat{k})$.

Several features of $\mathcal{G}(J, J'; k)$ merit comment. The quantities E and F depend on l and j , and not on the nature of the initial or final states. They are a set of numbers for electrons in the first ($j = l - \frac{1}{2}$) and second ($j = l + \frac{1}{2}$) part of the electron series. The properties of the initial and final states are solely contained in the reduced matrix element of $V^{(K')}$ which, since K' is odd, satisfies the identity (4.3). Classification of the states $n \geq 3$, and the coefficients of fractional parentage, have been tabulated by Bayman and Lande (1966). We have already noted that there are no dipole allowed transitions, since $V^{(1)}$ is diagonal with respect to the total angular momentum or transitions between states with different seniority numbers. The reduced matrix element of $V^{(K')}$ for $n = 2$ is given by (3.7). Application of our results to the case $l = 3$, reported in the following section, illustrates several features of the jj -coupling scheme and of the properties of $V^{(K')}$.

Because $V^{(1)}$ is diagonal with respect to the total angular momentum the leading term in equation (4.10) for $\mathcal{G}(J, J'; k)$ is $K' = 3$ when $J \neq J'$. The lowest order radial integral is therefore (j_2) and \mathcal{G} vanishes in the limit $k \rightarrow 0$. For $n = 1$ the cross section is obtained from the one-electron matrix elements provided in sections 3.1 and 3.2.

The j^n configuration in which $j = \frac{3}{2}$ is encountered in the description of d-electrons. The configuration $n = 4$ is a closed shell for which there is only one state, $J = 0$. For $n = 2$ the two states $J = 0, 2$ have different seniority quantum numbers. Hence, there are no inelastic events with the j^n -configurations in which $j = \frac{3}{2}$ and $n = 2, 4$. The one-particle cross sections are discussed in section 9.

7. Applications for $l = 3$

The f-electron series is discussed both to illustrate how some of the results work out in practice, and to make contact with available experimental spectroscopic data. To this end, table 3 contains a summary of the properties of the jj -coupling scheme for $l = 3$, illustrated for isolated tripositive rare earth ions. Ground states have a maximum J for j^n , which is $n\{j - (n - 1)/2\}$, with $j = \frac{5}{2}$ for f^1 - f^6 , and $j = \frac{7}{2}$ for f^7 - f^{13} . Complementary states, j^n and j^{2j+1-n} , have identical properties, e.g. f^2 and f^4 (f^8 and f^{12}) are complementary states with $j = \frac{5}{2}$ ($\frac{7}{2}$). There is one closed shell configuration, namely f^6 , which contains one state with $J = 0$. The seniority of the ground state is listed, together with the values of the total angular momentum (J') of states in j^n with the same ν because these are accessible from the ground state. For example, j^2 contains $J = 0, 2, 4$ in which $J = 0$ has $\nu = 0$ while $J = 2$ and 4 have $\nu = 2$. Since $V^{(K')}$ is diagonal with respect to ν the state $J' = 0$ is not accessible from the ground state $J = 4$. This result is borne out by the explicit result for the reduced matrix element of $V^{(K')}$ for $n = 2$ given in (3.7); with $J' = 0$ the $6j$ -symbol vanishes unless $J = K'$, but J and K' are required to be even and odd, respectively.

By examining table 3, the equivalent electron configurations j^n of immediate interest have $n = 2, 3, 4$, and 6 . For all but the closed shell, $n = \nu$ so the identity (4.3) does not bring any saving in labour. The values of $(\nu J || V^{(K')} || \nu J')$ required for $l = 3$ are provided in table 1.

8. Results for f^2

Here we report results for the elastic and inelastic neutron cross sections for f^2 , and compare them with corresponding results in the Russell-Saunders scheme. Within

Table 3. Total angular momentum and seniority quantum numbers for ground and excited states of tripositive rare earth ions. The ground and listed excited states for $n \geq 2$ have the same seniority quantum numbers, in view of the fact that $V^{(K')}$ for K' odd is diagonal with respect to ν .

	Ground state		Seniority	Excited states
$j = \frac{5}{2}$				
Ce ³⁺	f ¹	$J = \frac{5}{2}$	$n = 1$	$J' = \frac{7}{2}$
Pr ³⁺	f ²	$J = 4$	$n = 2$	$\nu = 2$ $J' = 2$
Nd ³⁺	f ³	$J = \frac{9}{2}$	$n = 3$	$\nu = 3$ $J' = \frac{3}{2}$
Pm ³⁺	f ⁴	$J = 4$	$n = 2$	$\nu = 2$ $J' = 2$
Sm ³⁺	f ⁵	$J = \frac{5}{2}$	$n = 1$	$(J' = \frac{7}{2})$
Eu ³⁺	f ⁶	$J = 0$	$n = 6$	
$j = \frac{7}{2}$				
Gd ³⁺	f ⁷	$J = \frac{7}{2}$	$n = 1$	$(J' = \frac{5}{2})$
Tb ³⁺	f ⁸	$J = 6$	$n = 2$	$\nu = 2$ $J' = 2, 4$
Dy ³⁺	f ⁹	$J = \frac{15}{2}$	$n = 3$	$\nu = 3$ $J' = \frac{3}{2}, \frac{5}{2}, \frac{9}{2}, \frac{11}{2}$
Ho ³⁺	f ¹⁰	$J = 8$	$n = 4$	$\nu = 4$ $J' = 2, 4, 5$
Er ³⁺	f ¹¹	$J = \frac{15}{2}$	$n = 3$	$\nu = 3$ $J' = \frac{3}{2}, \frac{5}{2}, \frac{9}{2}, \frac{11}{2}$
Tm ³⁺	f ¹²	$J = 6$	$n = 2$	$\nu = 2$ $J' = 2, 4$
Yb ³⁺	f ¹³	$J = \frac{7}{2}$	$n = 1$	$J' = \frac{5}{2}$

the jj -coupling scheme the results for j^2 apply also to j^{2j-1} . The final topic in the section is scattering from a state of two non-equivalent electrons ($j'j$) with $j' \neq j$ to a state of two equivalent electrons j^2 . We may note here that the amplitude for the event $j^2 \rightarrow j'^2$, $j \neq j'$, is zero.

8.1. Form factors

The paramagnetic and saturated ion form factors for elastic scattering are conveniently expressed in terms of the quantities $H(K')$ defined in (5.3). Armed with these quantities the paramagnetic and saturated ion form factors follow from (5.1) and (5.9) and from (5.8), respectively.

For $j = \frac{5}{2}$, $n = 2$, $J = J' = 4$ we obtain the following results from (3.7), (3.15), (3.16), (4.7) and (4.8) or, alternatively, table 1

$$\begin{aligned}
 H(1) &= \frac{12}{7} \left(\langle j_0 \rangle + \frac{8}{5} \langle j_2 \rangle \right) \\
 H(3) &= -\frac{3}{7} \left(\frac{1}{5} \right)^{3/2} \left(\langle j_2 \rangle + \frac{10}{3} \langle j_4 \rangle \right)
 \end{aligned} \tag{8.1}$$

and

$$H(5) = -\frac{10}{693} (\langle j_4 \rangle + 12 \langle j_6 \rangle).$$

Using these in (5.1) and (5.9), together with the Landé factor $g = \frac{6}{7}$, the paramagnetic form factor $\mathcal{F}(k)$ is obtained from

$$\mathcal{F}^2(k) = \left(\langle j_0 \rangle + \frac{8}{5} \langle j_2 \rangle \right)^2 + \frac{11}{(70)^2} \left(\langle j_2 \rangle + \frac{10}{3} \langle j_4 \rangle \right)^2 + \frac{13}{(66)^2} (\langle j_4 \rangle + 12 \langle j_6 \rangle)^2 \tag{8.2}$$

and it is displayed in figure 1. The corresponding quantity for a saturated ion is obtained from the appropriate structure factor averaged over the directions of \mathbf{k} and the result is given in (5.8). A comparison of expressions (5.8) and (5.9) leads us to the conclusion that the two form factors in question are built from the same combinations of $\langle j_K \rangle$ which appear in $H(K')$, and that only the coefficients differ. In the present case, the spatial average of the saturated ion structure factor leads to a form factor obtained from (8.2) by replacing the coefficients of the second and third terms by $1/1400$ and $5/47916$, respectively.

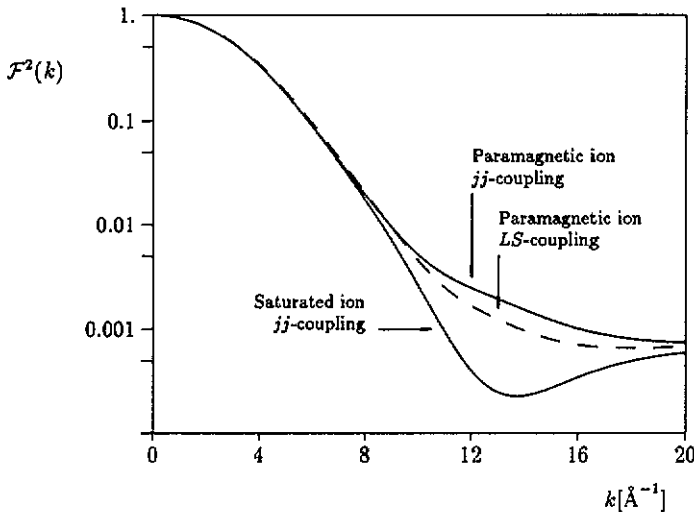


Figure 1. Paramagnetic and saturated ion form factors for f^2 calculated from (5.9) and (5.8), respectively, are displayed together with the corresponding paramagnetic form factor calculated with the Russell-Saunders scheme (Balcar and Lovesey 1989). In the absence of Dirac-Fock calculations for Pr^{3+} the figures are generated with radial integrals appropriate for U^{3+} . We anticipate that this choice of $\langle j_K \rangle$ is likely to affect amplitude values at large wavevectors ($\geq 12 \text{ \AA}^{-1}$) if at all.

Referring to figure 1, there is a significant difference in the numerical values of the two form factors at intermediate wavevectors. This is due largely to the fact that the coefficients of the second terms, which involve $(\langle j_2 \rangle + \frac{10}{3} \langle j_4 \rangle)^2$, differ by a factor 3.14. The paramagnetic form factor calculated according to the Russell-Saunders scheme is also included in figure 1, and we conclude that for f^2 there is a minimal difference in the elastic scattering cross sections for the two coupling schemes. A larger difference is revealed in the inelastic cross sections, as will be seen in the following sub-section.

8.2. Inelastic structure factor j^2

Just as in the preceding example of elastic scattering, the main building blocks for the scattering amplitude are the reduced matrix elements $(\nu J || V^{(K')} || \nu' J')$ and $E(K' - 1, K')$ and $F(K' - 1, K')$. Values for these quantities for $n = 2$, $j = \frac{5}{2}$, $J = 4$, $J' = 2$ can be obtained from (3.7), (4.7) and (4.8) or directly from tables 1 and 2.

Here we give the analytic expression for the total intensity for the transition $J = 4$, $J' = 2$ averaged over all directions of the wavevector. The appropriate quantity is

provided in (4.10), and in the present case we find

$$\mathcal{G}(J, J'; k) = \left(\frac{180}{2401} \right) \left(\langle j_2 \rangle + \frac{10}{3} \langle j_4 \rangle \right)^2 + \left(\frac{25}{5929} \right) \left(\langle j_4 \rangle + 12 \langle j_6 \rangle \right)^2. \quad (8.3)$$

Notice that the radial integrals appear in (8.3) and the elastic form factors in precisely the same combinations, i.e. the elastic and inelastic structure factors are linear combinations of the square of the quantities $H(K')$ defined in (5.3). By comparing the appropriate formulae, it is seen that the weight of $H^2(K')$ in a particular expression is largely determined by the reduced matrix element $(\nu J \| V^{(K')} \| \nu' J')$.

With regard to (8.3), there is no term in $H(1)$ because $(\nu J \| V^{(K')} \| \nu' J')$ is diagonal with respect to the total angular momentum, cf (3.6). This result is a general feature of inelastic transitions within a term described by j^n . Put in slightly different words, n equivalent atomic electrons described by the jj -coupling scheme do not possess dipole allowed transitions. The latter occur in the LS -scheme, and in jj -coupling they are found in non-equivalent electron configurations, as we illustrate in the next example.

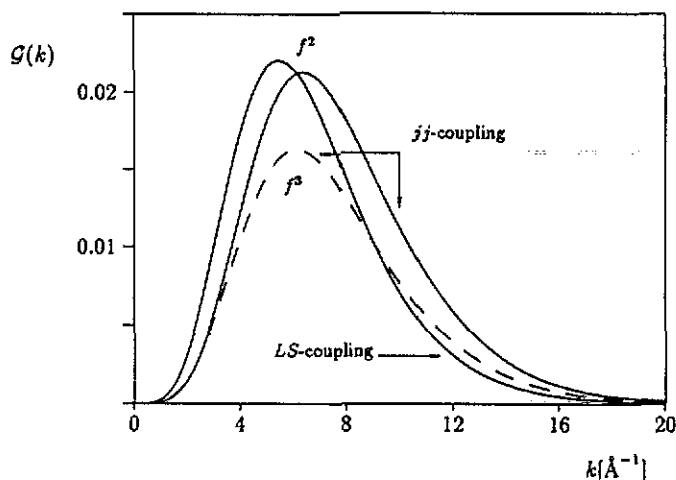


Figure 2. Inelastic structure factors calculated from (4.10) are shown for f^2 ($J = 4, J' = 2$) and f^3 ($J = \frac{9}{2}, J' = \frac{3}{2}$) together with the corresponding result for f^2 evaluated with the Russell-Saunders scheme (Balcar and Lovesey 1989). Note comments in figure 1.

The result (8.3) is displayed in figure 2 together with the corresponding quantity for LS -coupling. For this case ($j = \frac{5}{2}, J = 4, J' = 2$) a significant difference is found between the intensities evaluated by the two coupling schemes. The experimental resolution available today in scattering experiments is adequate to detect differences on the scale seen in figure 2.

As the final topic in this sub-section we compare results for f^2 and f^3 . This example is of interest in the interpretation of data for uranium compounds because there is often uncertainty as to the appropriate ionicity, indeed whether an integer value is appropriate. In view of this interest, we have included in figure 2 the intensity associated with the transition $j = \frac{5}{2}, J = \frac{9}{2}, J' = \frac{3}{2}$. To understand the difference

between the results for f^2 and f^3 we note the following. From the definition (4.10) of the total average intensity and the results for $(\nu J \| V^{(K')} \| \nu' J')$ for j^2 and j^3 provided in table 1 it is a simple matter to show that the intensity formula for j^3 , $J = \frac{3}{2}$, $J' = \frac{3}{2}$ is obtained from (8.3) by multiplying it by (4/5). Hence, the difference in the intensities for j^2 and j^3 apparent in figure 2 arise from two sources: (i) a simple reduction for j^3 with respect to j^2 of 20% which arises from differences in the coefficients in $\mathcal{G}(J, J'; k)$; and (ii) differences in the behaviour of the radial integrals for the atomic configurations j^2 and j^3 .

8.3. Non-equivalent electrons

To complete our discussion of scattering by an atom with two electrons described in jj -coupling we present the amplitude and cross section for the event $(j'j) \rightarrow j^2$ with $j \neq j'$. Let the two states have total angular momentum J' and J , respectively. A straightforward calculation reveals that the analogue of (3.3) is

$$(J' \| \chi^{K'} \| J) = (j' \| \chi^{K'} \| j) R(K') \quad (8.4)$$

where

$$R(K') = \sqrt{2}(-1)^{K'+j+j'} [J, J']^{1/2} \left\{ \begin{matrix} J' & K' & J \\ j & j & j' \end{matrix} \right\}.$$

The allowed J are restricted to the values $0, 2, \dots, (2j-1)$ because it is an antisymmetric state, whereas J' merely satisfies the triangle condition $|j-j'| \leq J' \leq (j+j')$ expressed in the $6j$ -symbol. Note that $R(K')$ plays a role similar to $V^{(K')}$ in the reduced matrix element for the configuration j^n , and there is a close resemblance between $R(K')$ and $V^{(K')}$ for $n=2$, given in equation (3.7). The phase factor in $R(K')$ reflects the ordering of the two one-electron states in the state J' constructed from $(j'j)$, i.e. there is a different phase for the state constructed from (jj') ; cf the discussion in section 3.2.

The inelastic structure factor averaged over all directions of k , defined in (2.11), is found to have the form

$$\begin{aligned} \mathcal{G}(J', J; k) = & 3 \sum_{K'=1} \{R(K') [A(K'-1, K') + B(K'-1, K')]\}^2 / (K'+1) \\ & + 3 \sum_{K=2} \{R(K) B(K, K)\}^2 / (2K+1). \end{aligned} \quad (8.5)$$

Here, the quantities $A(K, K')$ and $B(K, K')$ are proportional to the orbital and spin components of the one-electron reduced matrix element $(j' \| \chi^{K'} \| j)$ occurring in (8.4). The values of $A(K, K')$ and $B(K, K')$ for $l=2$ ($j' = \frac{3}{2}, j = \frac{5}{2}$) and $l=3$ ($j' = \frac{5}{2}, j = \frac{7}{2}$) are provided in table 4. For $l=3$ ($j' = \frac{7}{2}, j = \frac{5}{2}$), the values of $A(K'-1, K')$ and $B(K'-1, K')$ are obtained from the corresponding entries in table 4 by multiplying them by $(-\sqrt{3}/2)$, whereas the value of $B(K, K)$ is $(\sqrt{3}/2)$ times the corresponding entry. Returning to formula (8.5), let us note that the dependence of $\mathcal{G}(J', J; k)$ on the quantum numbers J', J is in $R(K')$. For given values of J', J the structure factor for $j' = \frac{7}{2}, j = \frac{5}{2}$ is the same as that for $j' = \frac{5}{2}, j = \frac{7}{2}$, evaluated with the aid of entries in table 4, apart from an overall multiplicative factor (3/4) which is just the ratio of the gyromagnetic ratios for the states $j = \frac{5}{2}$ and $\frac{7}{2}$, namely (6/7) and (8/7). When $l=2$, the structure factor for $j' = \frac{5}{2}, j = \frac{3}{2}$ is obtained from (8.5) using the entries in table 4 and multiplying the result by (2/3), the ratio of the gyromagnetic factors for $j' = \frac{3}{2}$ and $j = \frac{5}{2}$.

Table 4. The one-electron orbital and spin-reduced matrix elements $A(K, K')$ and $B(K, K')$ which occur in the structure factor (8.5) for non-equivalent electrons. The entries in the table are to be combined with the appropriate $\langle j_{K' \pm 1} \rangle$ as indicated by the indices for $A(K' - 1, K')$, $B(K' - 1, K')$, and with $\langle j_K \rangle$ for $B(K, K)$, respectively. For example $A(2, 3) = A(2, 3)_{2,4} \{ \langle j_2 \rangle + \langle j_4 \rangle \}$, $B(2, 3) = B(2, 3)_2 \langle j_2 \rangle + B(2, 3)_4 \langle j_4 \rangle$ and $B(2, 2) = B(2, 2)_2 \langle j_2 \rangle$.

$l = 2$			$l = 3$		
$j' = \frac{3}{2}$	$j = \frac{3}{2}$	$j = \frac{5}{2}$	$j' = \frac{3}{2}$	$j = \frac{5}{2}$	$j = \frac{7}{2}$
$A(0, 1)_{0,2}$	*011	011	$A(0, 1)_{0,2}$	*4211	2201
$A(2, 3)_{2,4}$	*4012	3112	$A(2, 3)_{2,4}$	*3112	4102
—	—	—	$A(4, 5)_{4,6}$	*3011, 2	2021, 2
$B(0, 1)_0$	011	*211	$B(0, 1)_0$	0211	*4201
$B(0, 1)_2$	*011	*211	$B(0, 1)_2$	*4211	*0201
$B(2, 2)_2$	0	*3131	$B(2, 2)_2$	0	*1311
$B(2, 3)_2$	2012	*7112	$B(2, 3)_2$	5112	*8302
$B(2, 3)_4$	*4212	*1312	$B(2, 3)_4$	*3112	*0102
$B(4, 4)_4$	—	*3311	$B(4, 4)_4$	0	*2201, 1
—	—	—	$B(4, 5)_4$	1011, 2	*4201, 2
—	—	—	$B(4, 5)_6$	*3031, 2	*0241, 2
—	—	—	$B(6, 6)_6$	—	*1321, 11

9. One-particle cross sections

We have chosen to write formulae (8.4) and (8.5) in such a convenient way as to cover the special cases d^1, d^9 and f^1, f^{13} as well, i.e. a single electron and a single hole. The structure factors for these two cases are obtained from (8.5) by setting $R(K') = 1$, and using the values of $A(K, K')$ and $B(K, K')$ provided in table 4; in this instance $J' = j'$ and $J = j$ with $j = l \pm \frac{1}{2}$. The inelastic structure factors for $\frac{5}{2} \rightarrow \frac{7}{2}$ and $\frac{7}{2} \rightarrow \frac{5}{2}$ ($l = 3$) are in the ratio $(6/7):(8/7)$, as explained in section 8.3. The results for $\frac{3}{2} \rightarrow \frac{5}{2}$ are also included in the table since they might be required for one-particle $l = 2$ events. In the case of elastic scattering ($j = j'$) the diagonal spin reduced matrix element ($K = K'$) is zero, and the structure factors defined in (4.10) and (8.5) have identical structures, as expected.

10. Discussion

Our comparative study of neutron scattering by electrons described by SL and jj -coupling schemes has revealed a number of significant differences. These might be seen in the interpretation of data to be signatures that indicate some evidence for the predominance of one scheme. We have presented specific results for f^2 by way of illustration. The elastic form factors are very similar, while the inelastic structure factors differ by an amount that is probably measurable.

Turning to general features it has been established that there are no dipole allowed transitions for j^n , $2 \leq n \leq (2j - 1)$. Also, the allowed transitions couple states with the same seniority. For one-particle states the only allowed single transition is of the dipole type, which means that the intensity is finite in the forward direction of scattering. Such events contribute to transitions to states with non-equivalent electrons, illustrated here by consideration of $(j', j) \rightarrow j^2$.

Acknowledgments

The paper was largely drafted while one of us (SWL) was a guest of the Aspen Institute of Physics (1990), and the Atominstitut of the Austrian Universities (1991). We thank Dr R Osborn for his interest in the work, and preparation of the figures. Petrine Villemoes participated in the initial stages of the investigation.

References

- Balcar E and Lovesey S W 1988 *J. Phys. C: Solid State Phys.* **21** L1127
— 1989 *Theory of Magnetic Neutron and Photon Scattering* (Oxford: Oxford University Press)
- Bayman B F and Lande A 1966 *Nucl. Phys.* **77** 1
- Brown P J, Piggis B N and Reynolds P A 1990 *J. Phys.: Condens. Matter* **2** 5297
- Edmonds A R 1974 *Angular Momentum in Quantum Mechanics* (Princeton, NJ: Princeton University Press)
- Judd B R 1963 *Operator Techniques in Atomic Spectroscopy* (New York: McGraw-Hill)
- Lindgren B 1989 *Institute of Physics Report* Uppsala University, Box 530, S-751 21 Uppsala, Sweden
- Lovesey S W 1987 *Theory of Electron Scattering from Condensed Matter* vol 2, 3rd impression (Oxford: Oxford University Press)
- Osborn R, Balcar E, Lovesey S W and Taylor A D 1991 *Physics and Chemistry of Rare Earths* vol 14, ed K A Gschneidner and L R Eyring (Amsterdam: North-Holland)
- Price D L and Skøld K 1986 *Neutron Scattering Methods of Experimental Physics* vols 23 A B C (New York: Academic)
- de-Shalit A and Talmi I 1963 *Nuclear Shell Theory* (New York: Academic)
- Stirling W G and McEwen K A 1987 *Methods of Experimental Physics* vol 23 part C, ed D L Price and K Skøld (Orlando: Academic)
- Stassis C and Deckman H W 1976 *Phys. Rev. B* **13** 4934