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Magnetic scattering of neutrons*

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The magnetic scattering of neutrons by an arbitrary system of particles has been examined by exploiting its similarity to the radiation problem in spectroscopy. It has been shown, in fact, that the magnetic scattering amplitude can be expressed in terms of the multipole moments of the scattering system. The number of multipoles, which contribute to the scattering amplitude, is limited by selection rules based on the symmetry properties of the states of the system, in particular, parity and angular momentum conservation. The formalism has been applied to the magnetic scattering of neutrons by an atom in the l^n electronic configuration. If the spin–other-orbit and orbit-orbit interactions in the atomic Hamiltonian can be neglected, only even-order electric and odd-order magnetic multipoles, whose order of multipolarity is less than or equal to $2l+1$, contribute to the scattering amplitude. In this case the calculation of the magnetic scattering amplitude is reduced to evaluating matrix elements of the Racah double tensors $W^{(0,k)}$ and $W^{(1,k')}$ (k' even). The former tensors are associated with the convection current and the latter with the spin magnetization contribution to the magnetic scattering amplitude. The calculation of the maxtrix elements of these tensors is simplified by selection rules based on the groups Sp($4l+2$), R($2l+1$), R(3), G_2 used in the classification of the atomic states. The contribution to the magnetic scattering amplitude of the convection current, associated with the spin-orbit and mass correction terms of the atomic Hamiltonian, has been examined in some detail.

I. INTRODUCTION

The magnetic scattering of neutrons by an atom has been the subject of many theoretical investigations. Following the original investigations of Bloch[1] and Schwinger,[2] Halpern and Johnson[3] examined the scattering by an atom with zero orbital magnetic moment. The general case of scattering by an atom with both spin and orbital magnetic moment was first examined by Trammell.[4] The matrix elements in the theory can be calculated by using either the traditional Condon and Shortley formalism or the more powerful techniques of Racah algebra.[5] However, the evaluation of the magnetic scattering amplitude by both methods requires rather involved calculations which offer little physical insight into the problem and which in some cases may even lead to erroneous results. It is the purpose of this paper to present a new treatment of the theory which makes possible the application of the techniques of modern spectroscopy to the magnetic scattering of neutrons. In this new formulation the calculation of the magnetic scattering by an atom is reduced to evaluating matrix elements of the generators of the groups used in the classification of the atomic states. The symmetry properties of the atomic states can then be used to considerably simplify the calculations.

We examine the magnetic scattering of neutrons by an arbitrary system of particles by exploiting the similarity of the problem with that of the interaction of radiation with the system. The magnetic scattering amplitude is determined by the Fourier transform of the transverse (to the scattering vector) component of the current density operator of the system. The Fourier transform of the transverse component of the current density operator can be simply related to the multipole moment operators of the scattering system. Thus the calculation of the magnetic scattering amplitude can be reduced to evaluating the transition matrix elements of the multipole moment operators. In any given problem the number of multipole operators used for the evaluation of the scattering amplitude is limited by selection rules based on parity and angular momentum conservation. The actual calculation of the matrix elements is a straightforward application of well-known techniques in atomic and nuclear spectroscopy.

The formalism has been applied to the magnetic scattering of slow neutrons by an atom in the l^n electronic configuration. If the two-particle momentum-dependent terms of the atomic Hamiltonian (such as the spin–other-orbit and the orbit-orbit interactions) can be neglected, the magnetic scattering amplitude can be expressed in terms of odd-order magnetic and even-order electric multipoles, respectively, whose order of multipolarity is less than or equal to $2l+1$. By relating the multipole operators to the Racah unit tensors, one can employ the symmetry properties of the atomic states to simplify the calculation of their matrix elements. The extensive tabulations of the reduced matrix elements of the Racah unit tensors can then be used for the practical evaluation of the magnetic scattering amplitude.

II. MAGNETIC SCATTERING OF NEUTRONS

We examine the scattering of neutrons arising from their electromagnetic interaction with an arbitrary system of charges. In the present work we will assume that the scattering system can be described by a nonrelativistic Hamiltonian. The neutron interaction with the system may be written

$$H_{\rm int} = -\frac{1}{c}\int \vec{j}(\vec{r})\cdot\vec{A}_n(\vec{r})\,d\vec{r} + \int \rho(\vec{r})\Phi_n(\vec{r})\,d\vec{r} - \frac{2\pi\hbar\mu_n}{Mc}\rho(\vec{r})\,, \qquad (1)$$

where $\vec{j}(\vec{r})$ and $\rho(\vec{r})$ denote the current and charge densities, respectively, of the system, and M is the neutron mass. The charge and current densities of the system are related by the continuity equation

$$\nabla\cdot\vec{j} = -\frac{\partial\rho}{\partial t} = -\frac{i}{\hbar}[H,\rho]\,, \qquad (2)$$

where H is the Hamiltonian of the system. For slow neutrons, the vector and scalar potentials due to the magnetic moment of the neutron are given by

$$\vec{A}_n(\vec{r}) = \frac{\vec{\mu}_n\times(\vec{r}-\vec{r}_n)}{|\vec{r}-\vec{r}_n|^3} \qquad (3)$$

and

$$\Phi_n(\vec{r}) = \frac{1}{c}\vec{\mu}_n\cdot\left(\vec{V}\times\frac{\vec{r}_n-\vec{r}}{|\vec{r}-\vec{r}_n|^3}\right). \qquad (4)$$

In these equations $\vec{r}_n$ is the position vector of the neutron, $\vec{V}$ is its velocity, $\vec{\mu}_n = \gamma\mu_N\vec{\sigma}$ is the magnetic moment of the neutron ($\gamma = -1.91$), and $\vec{\sigma}$ is the Pauli matrix. The first term in Eq. (1) is the interaction of the magnetic moment of the neutron with the convection and spin current of the system,[1-4] the second is the neutron-spin–neutron-orbit term arising from the coupling of the electric field of the moving neutron with the charge density of the system,[6] and the third is the Foldy term[7] due to the zitterbewegung motion of the anomalous magnetic moment of the neutron. In the present paper we will examine the contribution to the scattering cross section from the first term in Eq. (1). The corresponding scattering amplitude will be referred to as the magnetic scattering amplitude.

A. Magnetic scattering amplitude

We consider a scattering process in which the system undergoes a transition from some initial state of energy E_i to a final state of energy E_f, and the neutron is scattered from an initial state of wave vector $\vec{k}$ and energy E to a final state of wave vector $\vec{k}'$ and energy E'. The differential neutron scattering cross section in the first Born approximation can be written quite generally as

$$\frac{d^2\sigma}{d\Omega\,dE'} = (k'/k)\,|f(\vec{q})|^2\,\delta(E_i - E_f - \hbar\omega)\,, \qquad (5)$$

where

$$\vec{q} = \vec{k} - \vec{k}'\,, \qquad (6)$$

is the neutron scattering vector and $\hbar\omega = E' - E$ is the energy transferred to the neutron. For magnetic scattering, the scattering amplitude $f(\vec{q})$ is given by

$$f(\vec{q}) = \frac{M}{2\pi\hbar^2 c}\langle f|\int e^{i\vec{q}\cdot\vec{r}_n}\times\vec{j}(\vec{r})\cdot\frac{\vec{\mu}_n\times(\vec{r}-\vec{r}_n)}{|\vec{r}-\vec{r}_n|^3}\,d\vec{r}\,d\vec{r}_n|i\rangle\,.$$

After some simple manipulations, this equation may be written as

$$f(\vec{q}) = \frac{i\gamma r_0}{(2\pi)^3}\left(\frac{m}{e\hbar}\right)\vec{\sigma}\cdot\int d\vec{r}_n d\vec{q}'\,e^{i(\vec{q}-\vec{q}')\cdot\vec{r}_n}\times\frac{\vec{q}'}{q'^2}\times\langle f|\int \vec{j}(\vec{r})\,e^{i\vec{q}'\cdot\vec{r}}\,d\vec{r}|i\rangle\,, \qquad (7)$$

where m is the electronic mass, $r_0 = e^2/mc^2$ is the classical electron radius, and $e = -|e|$ denotes the electronic charge. This expression for the magnetic scattering amplitude can be considerably simplified if one assumes that the current density is independent of the neutron coordinates. This assumption is equivalent to neglecting terms in the interaction Hamiltonian which are quadratic, or of higher order, in $\vec{A}_n(\vec{r})$. With this assumption Eq. (7) reduces to

$$f(\vec{q}) = i(\gamma r_0)\,(m/e\hbar q)\vec{\sigma}\cdot(\hat{q}\times\vec{J}_\perp)\,, \qquad (8)$$

where $\vec{J}$ is the Fourier transform of the current density operator

$$\vec{J} = \langle f|\int \vec{j}(\vec{r})\,e^{i\vec{q}\cdot\vec{r}}\,d\vec{r}|i\rangle\,, \qquad (9)$$

and $\hat{q}$ is a unit vector along the scattering vector $\vec{q}$. It is seen that only $\vec{J}_\perp$, the component of $\vec{J}$ perpendicular to the scattering vector, contributes to the scattering amplitude. The neutron magnetic moment couples only to the transverse component of the current density. The magnetic scattering amplitude is sometimes written in a slightly different form by defining the dimensionless operator $\vec{\mathfrak{J}}_\perp$ as

$$\vec{\mathfrak{J}}_\perp = -i(m/e\hbar q)\hat{q}\times\vec{J}_\perp\,. \qquad (10)$$

With this definition the magnetic scattering amplitude may be written as

$$f(\vec{q}) = (|\gamma|\,r_0)\vec{\sigma}\cdot\vec{\mathfrak{J}}_\perp\,. \qquad (8')$$

The magnetic scattering amplitude has been related to the Fourier transform of the transverse component of the current density operator. The magnetic scattering of neutrons is thus similar to the problem of the interaction of polarized radiation with the system. Actually, the matrix elements of the spherical components of $\vec{J}_\perp$ (or $\vec{\mathfrak{J}}_\perp$) are proportional to those involved in the problem of the interaction of polarized radiation with the system. It is well known that these latter matrix elements are directly related to the multipole moments of the scattering system. Thus the magnetic scattering amplitude must also be simply related to the matrix elements of the multipole moments characterizing the scattering system.

B. Multipole moment expansion of the magnetic scattering amplitude

We showed that the magnetic scattering amplitude can be expressed in terms of the vectors $\vec{J}_\perp$ or $\vec{\mathfrak{J}}_\perp$ [Eq. (8) or (8′)]. These vectors will be determined in an arbitrary fixed coordinate system defined by the orthonormal vectors $\vec{\epsilon}_1$, $\vec{\epsilon}_2$, $\vec{\epsilon}_3$. However, instead of the Cartesian components of $\vec{J}_\perp$ (or $\vec{\mathfrak{J}}_\perp$), it is more convenient to use the components of these vectors along the spherical unit vectors $\vec{e}_p$ defined by

$$\vec{e}_p = -(p/\sqrt{2})(\vec{\epsilon}_1 + ip\vec{\epsilon}_2)\,, \quad p=\pm 1$$
$$\vec{e}_0 = \vec{\epsilon}_3\,.$$

The vectors $\vec{e}_p$ with $p=\pm 1$ are sometimes referred to as the helicity vectors associated with the direction $\vec{\epsilon}_3$. Since both $\vec{J}_\perp$ and $\vec{\mathfrak{J}}_\perp$ are transverse, they are completely defined by their components along the helicity vectors $\vec{e}_p^{\,\prime}$ associated with the scattering vector. First, the multipole moment expansion of these latter components will be determined. Then the multipole moment expansion of $\vec{J}_\perp$ and $\vec{\mathfrak{J}}_\perp$ in the fixed coordinate system will be obtained by a simple rotation of coordinates.

The components $J'_{\perp,p}$ of $\vec{J}_\perp$ along the helicity vectors $\vec{e}_p^{\,\prime}$ ($p=\pm 1$) associated with the scattering vector are

$$J'_{\perp,p} = \vec{J}_\perp \cdot \vec{e}_p^{\,\prime}$$
$$= \langle f|\int \vec{j}(\vec{r}) \cdot \vec{e}_p^{\,\prime} e^{i\vec{q}\cdot\vec{r}}\, d\vec{r}\,|i\rangle\,, \quad p=\pm 1\,. \tag{11}$$

The right-hand side of Eq. (11) is essentially the interaction Hamiltonian of polarized radiation with the system. The circularly polarized wave in the right-hand side of Eq. (11) can be expanded in multipole fields,[8]

$$\vec{e}_p^{\,\prime} e^{i\vec{q}\cdot\vec{r}} = -\frac{1}{\sqrt{2}} \sum_{k=1}^{\infty} \sum_{m=-k}^{+k} (p\vec{A}_{km}^{(m)} + \vec{A}_{km}^{(e)}) \times D^k_{mp}(\varphi, \vartheta, 0)\,, \tag{12}$$

where

$$\vec{A}_{km}^{(m)} = [k(k+1)]^{-1/2}\,\vec{l}\,\Phi_{km}(\vec{r})\,, \tag{13}$$

$$\vec{A}_{km}^{(e)} = (1/q)\,[k(k+1)]^{-1/2}\,\nabla\times\vec{l}\,\Phi_{km}(\vec{r})\,, \tag{14}$$

and

$$\Phi_{km}(\vec{r}) = i^k\,[4\pi(2k+1)]^{1/2} j_k(qr)\, Y_{km}(\hat{r})\,. \tag{15}$$

In these equations $\vec{l} = (1/i)\,\vec{r}\times\nabla$ is the angular momentum operator, $j_k(qr)$ is a spherical Bessel func-function, $D^k_{mp}(\varphi, \vartheta, 0)$ is a rotation matrix, and φ, ϑ are the polar angles of the scattering vector in the fixed coordinate system. The fields $\vec{A}_{km}^{(e)}$ and $\vec{A}_{km}^{(m)}$ are known as the electric and magnetic multipole components of the polarized wave and they have parities $(-)^{k+1}$ and $(-)^k$, respectively. Substituting Eq. (12) into Eq. (11)

$$J'_{\perp,p} = \langle f|\,-(2)^{-1/2}\sum_{k,m}\left(p\int \vec{A}_{km}^{(m)}\cdot\vec{j}\,d\vec{r} + \int \vec{A}_{km}^{(e)}\cdot\vec{j}\,d\vec{r}\right) D^k_{mp}(\varphi, \vartheta, 0)\,|i\rangle\,.$$

This equation can be written in the following compact form,

$$J'_{\perp,p} = -\left(\frac{e\hbar q}{m}\right)\sum_{k,m,\pi} p^{\pi} D^k_{mp}(\varphi, \vartheta, 0) \times\langle f|\,T_{km}^{(\pi)}\,|i\rangle\,, \quad p=\pm 1 \tag{16}$$

by introducing the dimensionless multipole operators[9]

$$T_{km}^{(e)} = \frac{1}{\sqrt{2}}\left(\frac{m}{e\hbar q}\right)\int \vec{A}_{km}^{(e)}\cdot\vec{j}\,d\vec{r} \tag{17a}$$

and

$$T_{km}^{(m)} = \frac{1}{\sqrt{2}}\left(\frac{m}{e\hbar q}\right)\int \vec{A}_{km}^{(m)}\cdot\vec{j}\,d\vec{r}\,. \tag{17b}$$

The operator $\vec{\mathfrak{J}}$ can also be expressed in terms of the multipole moment operators by using Eqs. (10) and (16),

$$\mathfrak{J}'_{\perp,p} = \left(\frac{m}{e\hbar q}\right) pJ'_{\perp,p} = -\sum_{k,m,\pi} p^{\pi+1} D^k_{mp}(\varphi, \vartheta, 0) \times\langle f|\,T_{km}^{(\pi)}\,|i\rangle\,, \quad p=\pm 1\,. \tag{18}$$

In Eqs. (16) and (18) π takes the values 0 and 1 for the electric and magnetic multipole operators, respectively. The superscripts (0) and (1) of the multipole operators then simply mean "electric" and "magnetic," respectively. The multipole operators [Eqs. (20)] are irreducible tensor operators of order k. Since the parity of $\vec{j}$ is $(-)$ the electric and magnetic multipole operators have parities $(-)^k$ and $(-)^{k+1}$, respectively.

We expressed the components of $\vec{J}_\perp$ and $\vec{\mathfrak{J}}_\perp$ along the helicity vectors associated with the scattering vector in terms of the multipole moment operators characterizing the scattering system. It is more convenient in some problems to use the spherical components of these vectors in the fixed coordinate system. These components can be easily obtained by a rotation of coordinates. The components of $\vec{J}_\perp$ in the fixed coordinate system are

$$J_{\perp,p}=\vec{J}_\perp\cdot\vec{e}_p=\sum_\sigma(-)^{p-\sigma}D^1_{-p,-\sigma}(\varphi,\vartheta,0)J'_{\perp,\sigma}\,,\quad p=0,\pm1 \tag{19}$$

where $J'_{\perp,\sigma}$ has been expressed in terms of the multipole moment operators [Eq. (16)]. A simple expression for $J_{\perp,p}$ can be obtained if one notes that

$$D^1_{-p,-\sigma}D^k_{m\sigma}=\sum_j(2j+1)\begin{pmatrix}k&1&j\\m&-p&p-m\end{pmatrix}\begin{pmatrix}k&1&j\\\sigma&-\sigma&0\end{pmatrix}\left(\frac{4\pi}{2j+1}\right)^{1/2}Y_{j,p-m}(\hat{q})$$

and

$$\sum_\sigma\sigma^\pi(-)^{-\sigma}\begin{pmatrix}k&1&j\\\sigma&-\sigma&0\end{pmatrix}=-[1-(-)^{k+j+\pi}]\begin{pmatrix}k&1&j\\1&-1&0\end{pmatrix}.$$

Substituting for $J'_{\perp,\sigma}$ in Eq. (19), one obtains

$$J_{\perp,p}=\left(\frac{e\hbar q}{m}\right)\sum_{\substack{k,m\\\pi,j}}(-)^p[1-(-)^{k+j+\pi}][4\pi(2j+1)]^{1/2}\begin{pmatrix}k&1&j\\m&-p&p-m\end{pmatrix}\begin{pmatrix}k&1&j\\1&-1&0\end{pmatrix}Y_{j,p-m}(\hat{q})\langle f|T^{(\pi)}_{km}|i\rangle\,,\quad p=0,\pm1\,. \tag{20}$$

The spherical components of $\vec{\mathcal{J}}_\perp$ in the fixed coordinate system are obtained by the same manipulations used in obtaining the corresponding components of $\vec{J}_\perp$:

$$\mathcal{J}_{\perp,p}=\sum_{\substack{k,m\\j,\pi}}(-)^p[1+(-)^{k+j+\pi}][4\pi(2j+1)]^{1/2}\begin{pmatrix}k&1&j\\m&-p&p-m\end{pmatrix}\begin{pmatrix}k&1&j\\1&-1&0\end{pmatrix}Y_{j,p-m}(\hat{q})\langle f|T^{(\pi)}_{km}|i\rangle\,,\quad p=0,\pm1\,. \tag{21}$$

Using Eqs. (20) and (21) the vectors $\vec{J}_\perp$ and $\vec{\mathcal{J}}_\perp$ can be written in a form which exhibits explicitly their transverse character. Let us consider first

$$\vec{J}_\perp=\sum_p(-)^pJ_{\perp,p}\vec{e}_{-p}\,,$$

with $J_{\perp,p}$ given by Eq. (20). The summations over π, j, and p can be easily performed if one notes that for $\pi=0$ (electric multipoles) j can take only the values $k\pm1$, and for $\pi=1$ (magnetic multipoles) j must be equal to k. The result is

$$\vec{J}_\perp=\left(\frac{e\hbar q}{m}\right)\sum_{k,m}\left(\frac{8\pi}{2k+1}\right)^{1/2}\{\vec{X}^*_{km}(\hat{q})\langle f|T^{(m)}_{km}|i\rangle-i[\hat{q}\times\vec{X}^*_{km}(\hat{q})]\langle f|T^{(e)}_{km}|i\rangle\}\,, \tag{22}$$

where $\vec{X}_{km}(\hat{q})$ is a vector spherical harmonic.[10] In deriving Eq. (22) we used the definition of the vector spherical harmonics and the following identity,

$$\hat{q}\times\vec{X}_{km}=i/(2k+1)^{1/2}[\sqrt{k}\,\vec{Y}^m_{k,k+1,1}+(k+1)^{1/2}\vec{Y}^m_{k,k-1,1}]\,,$$

where $\vec{Y}^m_{k,k+1,1}$, $\vec{Y}^m_{k,k-1,1}$ are vector spherical harmonics.[10] Using Eqs. (10) and (22), one can write the vector operator $\vec{\mathcal{J}}_\perp$ as

$$\vec{\mathcal{J}}_\perp=\sum_{k,m}\left(\frac{8\pi}{2k+1}\right)^{1/2}\{\vec{X}^*_{km}(\hat{q})\langle f|T^{(e)}_{km}|i\rangle-i[\hat{q}\times\vec{X}^*_{km}(\hat{q})]\langle f|T^{(m)}_{km}|i\rangle\}\,. \tag{23}$$

Equations (22) and (23) express the vectors $\vec{J}_\perp$ and $\vec{\mathcal{J}}_\perp$ in terms of their components along the transverse and mutually perpendicular vectors, $\vec{X}^*_{km}(\hat{q})$ and $[\hat{q}\times\vec{X}^*_{km}(\hat{q})]$.

We showed that the magnetic scattering amplitude can be expressed in terms of the matrix elements of the multipole operators characterizing the scattering system. In fact, by substituting Eq. (23) in Eq. (8′), one obtains

$$f(\vec{q})=(|\gamma|r_0)\vec{\sigma}\cdot\left[\sum_{k,m}\left(\frac{8\pi}{2k+1}\right)^{1/2}\{\vec{X}^*_{km}(\hat{q})\langle f|T^{(e)}_{km}|i\rangle-i[\hat{q}\times\vec{X}^*_{km}(\hat{q})]\langle f|T^{(m)}_{km}|i\rangle\}\right]\,. \tag{24}$$

In any given problem the number of multipole moment operators involved in the calculation of the magnetic scattering amplitude is limited by angular momentum and parity conservation. If the state of the scattering system is characterized by the total angular momentum J and its projection M along the z axis, then angular momentum conservation requires

$$|J_f-J_i|\le k\le J_f+J_i\,,\qquad M_f=M_i+m\,. \tag{25}$$

Parity conservation, on the other hand, requires k to be such that

$$P_f = (-)^k P_i \text{ for } \pi = 0 \text{ (electric multipoles)},$$
$$P_f = (-)^{k+1} P_i \text{ for } \pi = 1 \text{ (electric multipoles)}. \quad (26)$$

where P_i and P_f are the parities of the initial and final states, respectively, of the scattering system.

C. Multipole moment operators. Small-q approximation

The magnetic scattering amplitude has been related to the multipole moment operators characterizing the scattering system. We will examine here some of their properties which are of importance to the magnetic scattering of neutrons. It is well known that the magnetic and electric multipole operators are related to the magnetization and charge densities, respectively, of the system. Substituting for $\vec{A}_{km}^{(m)}$ [Eq. (13)] in Eq. (17b), one obtains

$$T_{km}^{(m)} = -2\left(\frac{m}{e\hbar q}\right) c i^{k-1}\left(\frac{2\pi(2k+1)}{k(k+1)}\right)^{1/2} \times \int d\vec{r}\, j_k(qr)\rho^{(m)}(\vec{r}) Y_{km}(\hat{r})$$
$$= -2\left(\frac{m}{e\hbar q}\right) c i^{k-1}\left(\frac{2\pi(2k+1)}{k(k+1)}\right)^{1/2} \times \int d\vec{r}\, \vec{M}(\vec{r}) \cdot \nabla[j_k(qr) Y_{km}(\hat{r})], \quad (27)$$

where

$$\vec{M}(\vec{r}) = (1/2c)\vec{r} \times \vec{j}(\vec{r}) \quad (28)$$

and

$$\rho^{(m)}(\vec{r}) = -\nabla \cdot \vec{M}(\vec{r}) \quad (29)$$

are the magnetization and the magnetic pole density, respectively, of the system. By substituting

$$\vec{A}_{km}^{(e)} = i^{k+1}\left(\frac{4\pi(2k+1)}{k(k+1)}\right)^{1/2}\left[q\vec{r}\, j_k(qr) Y_{km}(\hat{r}) + \frac{1}{q}\nabla\left(Y_{km}(\hat{r})\frac{d}{dr}[r j_k(qr)]\right)\right] \quad (30)$$

in Eq. (17a) and using the continuity equation [Eq. (2)], one obtains the following expression for the electric multipole operator:

$$T_{km}^{(e)} = i^{k+1}\left(\frac{m}{e\hbar q}\right)\left(\frac{2\pi(2k+1)}{k(k+1)}\right)^{1/2} \times \left(\frac{i(E_f - E_i)}{\hbar q}\int d\vec{r}\, Y_{km}(\hat{r})\rho(\vec{r})\frac{d}{dr}[r j_k(qr)] + q\int d\vec{r}\, j_k(qr) Y_{km}(\hat{r})\vec{r} \cdot \vec{j}(\vec{r})\right). \quad (31)$$

The current density $\vec{j}(\vec{r})$ consists of the convection current arising from the motion of the particles and the current associated with the spin magnetization of the system. The convection current is determined by the velocities of the particles and thus it depends explicitly on the momentum-dependent terms of the Hamiltonian of the system. Therefore, the electric and magnetic multipole operators [Eqs. (27), (28), and (31)] depend explicitly on the momentum-dependent terms of the Hamiltonian. Since the magnetic scattering amplitude is determined by the multipole operators, it will also depend explicitly on the momentum-dependent terms of the Hamiltonian. In most cases of experimental interest the dominant convection current contribution to the magnetic scattering amplitude arises from the kinetic-energy term of the Hamiltonian.

For a given current density, the multipole moment operators can be evaluated using Eqs. (27) and (31). A point-charge–point-magnetic-moment expression for the current density can be adopted if the effects associated with the internal structure of the particles can be neglected. With this assumption

$$\vec{j}(\vec{r}) = \vec{j}_c(\vec{r}) + \vec{j}_m(\vec{r}), \quad (32)$$

where the convection current $\vec{j}_c$ is

$$\vec{j}_c(\vec{r}) = \sum_i \frac{1}{2} e_i[\vec{v}_i\delta(\vec{r} - \vec{r}_i) + \delta(\vec{r} - \vec{r}_i)\vec{v}_i] \quad (32a)$$

and the spin magnetization current $\vec{j}_m$ is

$$\vec{j}_m(\vec{r}) = \sum_i g_i \frac{\hbar e_i}{2m_i}\nabla \times [\vec{s}_i\delta(\vec{r} - \vec{r}_i)]. \quad (32b)$$

The velocity operator of the ith particle $\vec{v}_i$ is defined by

$$\vec{v}_i = \frac{i}{\hbar}[H, \vec{r}_i] = \frac{\partial H}{\partial \vec{p}_i}, \quad (33)$$

where H is the Hamiltonian of the system. It is seen that the convection current is determined by the momentum dependent terms of the Hamiltonian. It is convenient to write the Hamiltonian in the following form:

$$H = H_0 + H_1 + H_2 + \cdots, \quad (34)$$

where H_0 consists of the kinetic energy and the momentum-independent terms of the Hamiltonian, H_1 consists of the one-particle momentum-dependent terms (except the kinetic energy), and H_2 denotes the two-particle momentum-dependent terms, etc. This separation of the Hamiltonian has been adopted because the dominant contribution to the convection current arises from the kinetic energy of the system, which is the only momentum-dependent term in H_0. Since

$$\vec{v}_i = \frac{\vec{p}_i}{m} + \sum_j \frac{\partial H_j}{\partial \vec{p}_i}, \quad j = 1, 2 \ldots \quad (35)$$

the multipole moment operators may be written as

$$T_{km}^{(\pi)} = T_{km}^{(\pi)}(0) + \sum_j T_{km}^{(\pi)}(j), \quad j = 1, 2 \ldots \; . \tag{36}$$

The first term in Eq. (36) is due to the magnetization current and the convection current associated with the kinetic energy of the system; the other terms arise from the convention current associated with the one-particle, two-particle, ... momentum-dependent terms of the Hamiltonian. Substituting the first term of Eq. (36) in Eqs. (31) and (27), one obtains

$$T_{km}^{(e)}(0) = i^{k+1}\left(\frac{m}{e\hbar q}\right)\left(\frac{2\pi(2k+1)}{k(k+1)}\right)^{1/2} \sum_i \left[\left\{\frac{i(E_f - E_i)}{\hbar q} e_i Y_{km}(\hat{r}_i)\frac{d}{dr}[r_i j_k(qr_i)] + \frac{qe_i}{2m_i}[j_k(qr_i)Y_{km}(\hat{r}_i)\vec{r}_i \cdot \vec{p}_i + \vec{p}_i \cdot \vec{r}_i j_k(qr_i)Y_{km}(\hat{r}_i)]\right\} - i\frac{g_i e_i \hbar q}{2m_i} \vec{s}_i \cdot \vec{l}_i [j_k(qr_i)Y_{km}(\hat{r}_i)]\right] \tag{37}$$

and

$$T_{km}^{(m)}(0) = i^{k}\left(\frac{m}{e\hbar q}\right)\left(\frac{2\pi(2k+1)}{k(k+1)}\right)^{1/2} \sum_i \frac{e_i\hbar}{m_i}\{i\nabla[j_k(qr_i)Y_{km}(\hat{r}_i)]\cdot\vec{l}_i + \tfrac{1}{2}g_i\vec{s}_i \cdot \nabla \times \vec{l}_i [j_k(qr_i)Y_{km}(\hat{r}_i)]\}\;. \tag{38}$$

The first term of Eq. (37) (in the large curly brackets) arises from the convection current associated with the kinetic energy and the second term from the spin magnetization current. A similar separation is seen in Eq. (38) for the magnetic multipole operator. The contributions to the multipole operators of the convection currents associated with the one-particle, two-particle, ... momentum-dependent terms of the Hamiltonian are

$$T_{km}^{(e)}(j) = i^{k+1}\left(\frac{m}{e\hbar q}\right)\left(\frac{2\pi(2k+1)}{k(k+1)}\right)^{1/2} \sum_i \frac{qe_i}{2}\left(j_k(qr_i)Y_{km}(\vec{r}_i)\vec{r}_i\cdot\frac{\partial H_j}{\partial\vec{p}_i} + \frac{\partial H_j}{\partial\vec{p}_i}\cdot\vec{r}_i j_k(qr_i)Y_{km}(\hat{r}_i)\right) \tag{39}$$

and

$$T_{km}^{(m)}(j) = i^{k}\left(\frac{m}{e\hbar q}\right)\left(\frac{2\pi(2k+1)}{k(k+1)}\right)^{1/2} \sum_i \frac{e_i}{2}\left(\vec{l}_i[j_k(qr_i)Y_{km}(\hat{r}_i)]\cdot\frac{\partial H_j}{\partial\vec{p}_i} + \frac{\partial H_j}{\partial\vec{p}_i}\cdot\vec{l}_i[j_k(qr_i)Y_{km}(\hat{r}_i)]\right). \tag{40}$$

Equations (37)–(40) are the point-charge–point-magnetic-moment expressions for the multipole operators. In most cases of experimental interest the dominant contribution to the magnetic scattering amplitude arises from the first term in Eq. (36). In this approximation $\vec{v}_i = \vec{p}_i/m$ and the multipole operators are determined by Eqs. (37) and (38).

If $qR \ll 1$, where R characterizes the size of the system, the multipole moment operators are proportional to the static multipole moments of the system. In fact, it is easily seen [Eqs. (27) and (31)] that in this limit

$$T_{km}^{(m)} = -\left(\frac{m}{e\hbar q}\right)\frac{2ci^{k-1}q^k}{(2k+1)!!}\left(\frac{2\pi(2k+1)}{k(k+1)}\right)^{1/2}\int d\vec{r}\, r^k \rho^{(m)}(\vec{r})Y_{km}(\hat{r}), \quad qr \ll 1 \tag{41}$$

and

$$T_{km}^{(e)} = -\left(\frac{m}{e\hbar q}\right)\frac{i^k q^k}{(2k+1)!!}\left(\frac{2\pi(2k+1)(k+1)}{k}\right)^{1/2}\frac{E_f - E_i}{\hbar q}\int d\vec{r}\, r^k \rho(\vec{r})Y_{km}(\hat{r}), \quad qr \ll 1. \tag{42}$$

In deriving these equations we used the small-argument expression for the spherical Bessel function

$$j_k(qr) \simeq \frac{(qr)^k}{(2k+1)!!}, \quad qr \ll 1. \tag{43}$$

Note the close connection in this limit of the multipole operators with the static multipole moments given in the integrals in terms of the magnetic and charge densities of the system. Equation (42) is the mathematical statement of the well-known Siegert's theorem in spectroscopy: The electric multipole operators in the small-q approximation do not depend explicitly on the momentum-dependent terms of the Hamiltonian. The small-q expressions for the multipole operators [Eqs. (41) and (42)] can be used for most problems of nuclear and atomic spectroscopy, since the wavelengths involved in these problems are much larger than the linear dimensions of the systems. These expressions, on the other hand, are of limited applicability in the magnetic scattering problem, since the neutron wavelengths are usually of the order of 1 Å.

If the initial and final states of the system are of different parity only the electric dipole term [$k = 1$ in Eq. (42)] is of importance at small values of q. The magnetic scattering amplitude [Eq. (24)] at small values of q is then independent of the magnitude of the scattering vector and is proportional to the transition matrix element of the electric dipole moment of the system. If the magnetic scattering of neutrons occurs without change in the parity of the scattering system only the magnetic dipole term [$k=1$ in Eq. (41)] is of importance at small values of q. In this case the magnetic scattering amplitude is independent of q and proportional to the transition matrix element of the magnetic moment of the system.

III. MAGNETIC SCATTERING BY AN ATOM

In this section we will examine the magnetic scattering of neutrons by the electrons of a free atom or ion. It has been shown in Sec. II that the magnetic scattering amplitude can be expressed in terms of the multipole moment operators. Adopting the point-particle model for the atomic electrons, these operators are given by Eqs. (37)–(40). We will examine the scattering by an atom in the l^n electronic configuration, and we will assume that the energy of the neutron is sufficiently low so that only transitions within the l^n configuration are possible. In this case, by parity conservation [Eq. (26)], only even-order electric and odd-order magnetic multipoles contribute to the magnetic scattering amplitude. The calculation of the magnetic scattering amplitude is thus reduced to evaluating matrix elements between states $|JM\rangle$ of the atom, of even-order electric and odd-order magnetic multipoles. These states are, in general, determined from Russel-Saunders states $|\theta JM\rangle$ of the l^n configuration:

$$|JM\rangle = \sum_\theta a(\theta)\,|\theta JM\rangle\,, \tag{44}$$

where $\theta=\alpha SL$ and α stands for additional quantum numbers needed to specify the state. Assuming that the $a(\theta)$'s are known, the calculation of the magnetic scattering amplitude is reduced to evaluating matrix elements of the form $\langle\theta JM|T_{km}^{(\pi)} \times|\theta' J' M'\rangle$, where the multipole operators are given by Eqs. (37)–(40), and the order of multipolarity k must be even for electric multipoles ($\pi=0$) and odd for magnetic multipoles ($\pi=1$). In order to take full advantage of the symmetry properties of the atomic states, the multipole operators will be expressed in terms of the Racah unit tensors $W^{(k',k'')}$ defined in the Appendix. The operators $W^{(k',k'')}$ are double tensors having rank k' in the spin space of the atom (defined by $\vec{S}=\sum_i\vec{s}_i$), rank k'' in the orbital space of the atom (defined by $\vec{L}=\sum_i\vec{l}_i$), and they form the basic building blocks for describing the symmetry properties of atomic interactions. By using the Racah unit tensors the calculation of the matrix elements of the multipole operators is separated into (a) evaluation of radial matrix elements, and (b) evaluation of angular matrix elements. The radial matrix elements can be calculated using radial wavefunctions obtained from some type of Hartree-Fock calculation. The angular matrix elements are determined by the matrix elements of the Racah unit tensors. The calculation of these latter matrix elements is considerably simplified by symmetry properties, since the double tensors $W^{(k',k'')}$ are the generators of the group $U(4l+2)$ used in the classification of the atomic states. The availability of extensive tabulations of these matrix elements considerably simplifies the calculation of the magnetic scattering amplitude.

In the extreme nonrelativistic limit the only momentum-dependent term in the atomic Hamiltonian is the electronic kinetic energy. In this case the multipole moment operators are simply given by Eqs. (37) and (38). The extreme nonrelativistic limit is an excellent approximation for most problems in neutron scattering. This is because the momentum-dependent terms in the atomic Hamiltonian, other than the kinetic energy, are relativistic corrections of the order of $(v/c)^2$, the electronic kinetic energy. In light atoms the contribution of these relativistic terms to the electronic convection current is negligible in comparison to that of the electronic kinetic energy. In heavy atoms, however, the contribution to the electronic convection current of the spin-orbit interaction and the mass correction terms can be of some importance. In this section we first evaluate the magnetic scattering amplitude in the extreme nonrelativistic limit and then we include the relativistic corrections due to the spin-orbit and mass correction terms. The calculations are a simple application of well-known techniques in atomic spectroscopy. [11]

The multipole operators in the extreme nonrelativistic limit [Eqs. (37) and (38)] and the multipole operators associated with the spin-orbit and mass correction terms are one-particle operators. These operators can be easily expressed in terms of the Racah unit tensors if one notes that they are linear combinations of one-particle operators of the form

$$\sum_i [a_i^{k'}\times b_i^{k''}]^k_m\,, \tag{45}$$

where $a_i^{k'}$ and $b_i^{k''}$ are one-electron tensor operators (of rank k' and k'') acting on the spin and orbital coordinates, respectively, of the ith electron, and $[a_i^{k'}\times b_i^{k''}]^k$ denotes the tensor of rank k formed by their tensor product. The matrix elements of these operators can be written in the following form[11]:

$$\left(\theta JM\left|\sum_i [a_i^{k'}\times b_i^{k'}]_m^k\right|\theta' J' M'\right)=[k',k'']^{-1/2}(s||a^{k'}||s)(l||b^{k''}||l)(\theta JM|W_m^{(k',k'')k}|\theta' J' M'), \tag{46}$$

where $[k', k'']$ stands for $[(2k'+1)(2k''+1)]$ and $W_m^{(k',k'')k}$, the mth component of a tensor of rank k, is defined in the Appendix. The tensor $W^{(k',k'')k}$ is a unit tensor of rank k' and k'' in the spin and orbital space of the atom, respectively. The matrix elements of these tensors are simply related to the reduced matrix elements of the Racah unit tensors by[11]

$$(\theta JM|W_m^{(k',k'')k}|\theta' J' M')=(-)^{J-M}[J,k,J']^{1/2}\begin{pmatrix} J & k & J'\\ -M & m & M'\end{pmatrix}\begin{Bmatrix} S & S' & k'\\ L & L' & k''\\ J & J' & k\end{Bmatrix}(\theta||W^{(k',k'')}||\theta'). \tag{47}$$

In the following calculations the multipole operators are first expressed in terms of tensors of the form given by Eq. (45), then their matrix elements are related to the reduced matrix elements of the Racah tensors by using Eqs. (46) and (47). Since there is little point in reporting details of algebraic manipulations, only an outline of the calculations is given in this paper.

A. Extreme nonrelativistic approximation

In the extreme nonrelativistic approximation the multipole operators are determined by Eqs. (37) and (38). We recall that, by parity, only even-order electric and odd-order magnetic multipole operators contribute to the scattering amplitude. In addition, the order of multipolarity of these multipoles must be less or equal to $2l+1$, since these operators are one-particle operators and we are considering only atomic transitions within the l^n electronic configuration.

The operators $T_{km}^{(e)}(0)$ and $T_{km}^{(m)}(0)$ [Eqs. (37) and (38)] can be expressed in terms of operators of the form given by Eq. (45) if one notes that

$$\vec{s}\cdot\vec{l}(j_kY_{km})=-[k(k+1)]^{1/2}(\vec{s}\times j_kY_k)_m^k\ ,$$

$$\nabla(j_kY_{km})\cdot\vec{l}=q\left[\left(\frac{k+1}{2k+1}\right)^{1/2}[j_{k+1}Y_{k+1}\times\vec{l}]_m^k+\left(\frac{k}{2k+1}\right)^{1/2}[j_{k-1}Y_{k-1}\times\vec{l}]_m^k\right],$$

and

$$\vec{s}\cdot\nabla\times[\vec{l}(j_kY_{km})]=iq[k(k+1)]^{1/2}\left[-\left(\frac{k}{2k+1}\right)^{1/2}[\vec{s}\times j_{k+1}Y_{k+1}]_m^k+\left(\frac{k+1}{2k+1}\right)^{1/2}[\vec{s}\times j_{k-1}Y_{k-1}]_m^k\right].$$

Using these relations and Eq. (46) one obtains

$$(\theta JM|T_{km}^{(m)}(0)|\theta' J' M')=i^{k+1}\left(R_0(k)(\theta JM|W_m^{(0,k)k}|\theta' J' M')+\sum_{k'=k\pm1}R_1(k',k)(\theta JM|W_m^{(1,k')k}|\theta' J' M')\right)\quad k=1,3\ldots 2l+1 \tag{48}$$

and

$$(\theta JM|T_{km}^{(e)}(0)|\theta' J' M')=i^{k+1}[R_2(k)(\theta JM|W_m^{(0,k)k}|\theta' J' M')+R_1(k,k)(\theta JM|W_m^{(1,k)k}|\theta' J' M')],\quad k=2,4\ldots 2l. \tag{49}$$

It is seen that the matrix elements are naturally separated into radial and angular matrix elements. The angular matrix elements are given in terms of the reduced matrix elements of the Racah unit tensors by Eq. (47). Note also in these expressions, the separation of the orbital and spin magnetization contributions to the multipole operators. As expected, the orbital contribution is expressed in terms of the $W^{(0,k)}$, and the spin magnetization contribution is given in terms of the $W^{(1,k')}$ (or $W^{(1,k)}$) Racah unit tensors.

The radial matrix elements in Eqs. (51) and (52) can be calculated using radial wave functions obtained from some type of Hartree-Fock calculation. If one denotes by $f(r)$ and $f'(r)$ the single-electron radial wave functions of the states $|\theta JM\rangle$ and $|\theta' J' M'\rangle$, respectively, the radial matrix elements can be written as

$$R_0(k)=(-)^{k+l}(2l+1)\left(\frac{l(l+1)(2l+1)(2k+3)}{k}\right)^{1/2}\begin{pmatrix} l & k+1 & l\\ 0 & 0 & 0\end{pmatrix}\begin{Bmatrix} k+1 & 1 & k\\ l & l & l\end{Bmatrix}(\bar{j}_{k+1}+\bar{j}_{k-1}), \tag{50}$$

$$R_1(k',k)=(-)^l i^{k-k'-1}(2l+1)\left(\frac{(2k+1)(2k'+1)}{2}\right)^{1/2}\begin{pmatrix}k' & 1 & k\\ 0 & 1 & -1\end{pmatrix}\begin{pmatrix}l & k' & l\\ 0 & 0 & 0\end{pmatrix}\bar{j}_{k'}, \tag{51}$$

and

$$R_2(k)=\frac{(-)^l}{2i}(2l+1)\left(\frac{2k+1}{k(k+1)}\right)^{1/2}\begin{pmatrix}l & k & l\\ 0 & 0 & 0\end{pmatrix}\bar{R}_2. \tag{52}$$

In these equations

$$\bar{j}_k=\int_0^\infty r^2 f(r)f'(r)j_k(qr)\,dr \tag{53}$$

and

$$\bar{R}_2=\frac{E_i-E_f}{\hbar^2q^2/2m}\int_0^\infty r^2 f(r)f'(r)\frac{d[rj_k(qr)]}{dr}\,dr+\int_0^\infty r^3 j_k(qr)\left(f\frac{df'}{dr}-f'\frac{df}{dr}\right)dr. \tag{54}$$

The integrals $\bar{j}_k$ for transition-metal and rare-earth atoms have been calculated[12] using Hartree-Fock radial wave functions.

The evaluation of the angular matrix elements is simplified by symmetry considerations. The atomic states of the l^n configuration can be classified according to the irreducible representations of the groups $Sp(4l+2)$, $R(2l+1)$, $R(3)$, and in the case of f electrons the special group G_2. The angular matrix elements in Eqs. (48) and (49) are matrix elements of the tensors $W^{(0,k)k}$ and $W^{(1,k')k}$, where k' is an even integer. The selection rules based on R(3) are the familiar selection rules on angular momentum. The matrix elements of $W^{(0,k)k}$ vanish unless $|J-J'|\le k\le J+J'$, $|L-L'|\le k\le L+L'$ and $S'=S$; similarly the matrix elements of $W^{(1,k')k}$ vanish unless $|J-J'|\le k\le J+J'$, $|L-L'|\le k' \le L+L'$ and $S'=S$, $S\pm 1$. The selection rules based on $Sp(4l+2)$ can be simply expressed by using the seniorities of the atomic states. The odd tensors $W^{(0,k)}$ (k odd) and $W^{(1,k')}$ are diagonal in seniority. The tensors $W^{(0,k)}$ (k even), on the other hand, can only link states differing in seniority by 0 or 2. The selection rules based on $R(2l+1)$ impose additional restrictions. The operators $W^{(0,k)}$ (k odd) can only connect states transforming according to the same representation W of $R(2l+1)$, because these operators are the generators of the group.

Regarding the matrix elements of $W^{(1,k')}$ and $W^{(0,k)}$ (k even), the selection rules based on $R(2l+1)$ cannot be so concisely stated. If one of these tensors transforms like the representation W of $R(2l+1)$, its matrix element between two states transforming like W' and W'' will vanish if the coefficient $C(W'W''W)$ in the decomposition of the Kronecker product $W'\times W''=\sum_W C(W'W''W)W$ vanishes. For instance, the selection rules based on R(5) (d electrons) and R(7) (f electrons) are easily obtained by using the available tables of $C(W'W''(20))$ and $C(W'W''(200))$, respectively, if one recalls that the tensors $W^{(1,k')}$ and $W^{(0,k)}$ (k even) transform like the representations (20) and (200) of the groups R(5) and R(7), respectively. In the case of f electrons the double tensor $W^{(0,5)}$ and $W^{(0,1)}$ form the generators of the special group G_2 and therefore can connect only states transforming according to the same representation U of G_2.

The nonvanishing reduced matrix elements of the double tensors $W^{(0,k)}$ and $W^{(1,k')}$ (k' even), needed for the evaluation of the magnetic scattering amplitude, can be calculated by means of the formula

$$(\theta\,||\,W^{(k',k'')}\,||\,\theta')=n[S,S',L,L',k',k'']^{1/2}\times\sum_{\bar\theta}(\theta\{|\bar\theta)(\theta'\{|\bar\theta)(-)^{\bar S+1/2+S+k'+\bar L+l+L+k''}\times\begin{Bmatrix}S & k' & S'\\ \frac12 & \bar S & \frac12\end{Bmatrix}\begin{Bmatrix}L & k'' & L'\\ l & \bar L & l\end{Bmatrix}, \tag{55}$$

where $\bar\theta=\bar\alpha\,\bar S\,\bar L$ defines a term of l^{n-1} and $(\theta\{|\bar\theta)$ is a coefficient of fractional parentage (cfp). Nielson and Koster[13] tabulated the cfp for states of the p^n, d^n, and f^n configurations. For most cases of practical interest the reduced matrix elements of the double tensors $W^{(0,k)}$ and $W^{(1,k')}$ have been tabulated. For the p^n, d^n, and f^n configurations with $n\le 2l+1$ the reduced matrix elements of $W^{(0,k)}$ can be obtained directly from the work of Nielson and Koster,[13] which tabulated the reduced matrix elements of $U^k=[2/(2k+1)]^{1/2}W^{(0,k)}$. If $n>2l+1$ the reduced matrix elements may be obtained from those with $n\le 2l+1$ by using the relation

$$(l^n vSL\,||\,W^{(k',k'')}\,||\,l^n v'S'L')=(-)^{k'+k''+1+(v-v')/2}\times(l^{4l+2-n}vSL\,||\,W^{(k',k'')}\,||\,l^{4l+2-n}v'S'L'), \tag{56}$$

where v and v' are the seniorities of the states. The same tables can be used to evaluate the reduced matrix elements of $W^{(1,k')}$ (k' even), since they are simply related[14] to those of the $W^{(0,k)}$ tensors. The reduced matrix elements of the double tensors $W^{(1,k')}$ have been tabulated by Karaziya *et al.*[15] for the p^n, d^n, f^2, f^3 and f^4 configurations.

The calculation of the magnetic scattering amplitude has been reduced to evaluating reduced matrix elements of the double tensors $W^{(0,k)}$ and $W^{(1,k')}$ (k' even). Denoting by $|J'M'\rangle$ and $|JM\rangle$ the initial and final states of the atom, respectively, the magnetic scattering amplitude can be written as [Eq. (44)]

$$f(\vec{q}) = \sum_{\theta,\theta'} a^*(\theta)a(\theta')f^{\theta JM}_{\theta' J' M'}(\vec{q}), \tag{57}$$

where $f^{\theta JM}_{\theta' J' M'}(\vec{q})$, the magnetic scattering amplitude for the transition $\theta'J'M' \to \theta JM$, is

$$f^{\theta JM}_{\theta' J' M'}(\vec{q}) = (|\gamma| r_0)\vec{\sigma}\cdot\sum_{k,m} i^{k+1}\left(\frac{8\pi}{2k+1}\right)^{1/2}\Big[\vec{X}^*_{km}(\hat{q})[R_2(k)(\theta JM|W^{(0,k)k}|\theta'J'M') + R_1(k,k)(\theta JM|W^{(1,k)k}_m|\theta'J'M')]$$
$$- i[\hat{q}\times\vec{X}^*_{km}(\hat{q})]\Big(R_0(k)(\theta JM|W^{(0,k)k}_m|\theta'J'M') + \sum_{k'=k\pm1} R_1(k',k)(\theta JM|W^{(1,k')k}_m|\theta'J'M')\Big)\Big]. \tag{58}$$

Equation (58) has been obtained by substituting Eqs. (48) and (49) for the matrix elements of the multipole operators in Eq. (24). In the first term in the large square brackets, the electric multipole term, $k = 2, 4, \ldots, 2l$ and in the second, the magnetic multipole contribution $k = 1, 3, \ldots, 2l+1$. The electric multipole contribution was not included in previous formulations of the theory.[4,5] The magnetic amplitude for elastic scattering can be considerably simplified if the state of the atom or ion can be approximately characterized by a single Russell-Saunders state $|\theta JM\rangle$. In this case the electric multipole contribution vanishes and only the $m=0$ components of the magnetic multipoles contribute to the scattering amplitude. Therefore,

$$f^{\theta JM}_{\theta JM}(\hat{q}) = (|\gamma| r_0)\vec{\sigma}\cdot\sum_{k} i^k\left(\frac{8\pi}{2k+1}\right)^{1/2}[\hat{q}\times\vec{X}^*_{k0}(\hat{q})]\Big(R_0(k)(\theta JM|W^{(0,k)k}_0|\theta JM) + \sum_{k'=k\pm1} R_1(k',k)(\theta JM|W^{(1,k')k}_0|\theta JM)\Big), \tag{59}$$

where $k = 1, 3, 5, \ldots 2l+1$. Equation (59) is a good approximation for the magnetic scattering amplitude, in the case of elastic scattering by rare-earth ions in their ground state.

B. Relativistic corrections

We calculated the magnetic scattering amplitude [Eqs. (57) and (58)] by assuming that the only momentum dependent term in the atomic Hamiltonian is the electronic kinetic energy. This is a good approximation for light atoms, since the other momentum dependent terms in the Hamiltonian are relativistic corrections of the order of $(v/c)^2$, the kinetic-energy term. In heavy atoms, however, the contribution to the scattering amplitude of the spin-orbit and mass correction terms may be of some importance. The contributions of these terms to the multipole moment operators can be calculated by using Eqs. (39) and (40) with

$$H_1 = \sum_i \xi(r_i)\vec{l}_i\cdot\vec{s}_i - \frac{p_i^4}{8m^3c^2}, \tag{60}$$

$$\vec{v}_i = \frac{\partial H_1}{\partial \vec{p}_i} = \frac{1}{\hbar}\xi(r_i)(\vec{s}_i\times\vec{r}_i) - \frac{p_i^2}{2m^3c^2}\vec{p}_i. \tag{61}$$

The selection rules are the same as in the extreme nonrelativistic calculations: $k = 1, 3\ldots, 2l+1$ for magnetic multipoles and $k = 2, 4\ldots, 2l$ for electric multipoles. The operators $T^{(m)}_{km}(1)$ and $T^{(e)}_{km}(1)$ can be expressed in terms of operators of the form given by Eq. (45) if one notes that

$$\left(\frac{2k+1}{k(k+1)}\right)^{1/2}\vec{s}\{\xi(r)\vec{r}\times\vec{l}[j_k(qr)Y_{km}(\hat{r})]\} = i\{\sqrt{k}[\vec{s}\times r\xi(r)j_k(qr)Y_{k+1}(\hat{r})]^k_m + (k+1)^{1/2}[\vec{s}\times r\xi(r)j_k(qr)Y_{k-1}(\hat{r})]^k_m\}. \tag{62}$$

Using Eqs. (60)–(62), the matrix elements of $T^{(m)}_{km}$ and $T^{(e)}_{km}(1)$, Eqs. (39) and (40), can be expressed in terms of the Racah tensors:

$$T^{(m)}_{km}(1) = i^{k+1}\Big(\sum_{k'=k\pm1} R_3(k',k)\langle\theta JM|W^{(1,k')k}_m|\theta'J'M'\rangle + R_4(k)\langle\theta JM|W^{(0,k)k}_m|\theta'J'M'\rangle\Big),\quad k = 1, 3, \ldots, 2l+1 \tag{63}$$

and

$$T^{(e)}_{km}(1) = i^{k+1}R_5(k)\langle\theta JM|W^{(0,k)k}_m|\theta'J'M'\rangle,\quad k = 2, 4, \ldots, 2l. \tag{64}$$

The radial matrix elements in these equations are

$$R_3(k',k)=(-)^l(2l+1)\left(\frac{(2k'+1)}{2(2k+1)}\right)^{1/2}\begin{pmatrix}k' & 1 & k\\ 0 & 1 & -1\end{pmatrix}\begin{pmatrix}l & k' & l\\ 0 & 0 & 0\end{pmatrix}\overline{R}_3(k),$$

$$\overline{R}_3(k)=\frac{m}{\hbar^2}\int r^4(j_{k+1}+j_{k-1})\,\xi(r)f(r)f'(r)\,dr;$$

$$R_4(k)=(-)^{k+l}(2l+1)\left(\frac{l(l+1)(2l+1)(2k+3)}{k}\right)^{1/2}\begin{pmatrix}l & k+1 & l\\ 0 & 0 & 0\end{pmatrix}\begin{Bmatrix}k+1 & 1 & k\\ l & l & l\end{Bmatrix}\overline{R}_4(k),$$

$$\overline{R}_4(k)=\frac{\hbar^2}{4m^2c^2}\int r^2\,dr\,(j_{k+1}+j_{k-1})\left[f\left(\nabla^2 f'-\frac{l(l+1)}{r^2}f'\right)+f'\left(\nabla^2 f-\frac{l(l+1)}{r^2}f\right)\right]; \tag{65}$$

and

$$R_5(k)=\frac{(-)^l}{i}(2l+1)\left(\frac{2k+1}{k(k+1)}\right)^{1/2}\begin{pmatrix}l & k & l\\ 0 & 0 & 0\end{pmatrix}\overline{R}_5(k),$$

$$\overline{R}_5(k)=\frac{\hbar^2}{4m^2c^2}\int r^3\,dr\,j_k(qr)\left[f\frac{d}{dr}(\nabla^2 f')-f'\frac{d}{dr}(\nabla^2 f)-\frac{l(l+1)}{r^2}\left(f\frac{df'}{dr}-f'\frac{df}{dr}\right)\right].$$

The angular matrix elements in Eqs. (63) and (64) can be related to the reduced matrix elements of the double tensors by using Eq. (50). It is seen that the calculation of the contribution to the multipole moments of the spin-orbit and mass correction terms is again reduced to evaluating reduced matrix elements of double tensors $W^{(0,k)}$ and $W^{(1,k')}$ (k' even). The evaluation of these reduced matrix elements is performed as indicated in the discussion of the extreme nonrelativistic approximation.

An order-of-magnitude calculation shows that in the elastic scattering by rare-earth ions the contributions of the spin-orbit and mass correction terms to the magnetic scattering amplitude are comparable, and of the order of a few parts per thousand of the total amplitude. These relativistic effects can be of importance in the investigation of the conduction-electron polarization effects in the rare-earth metals, since the conduction-electron contribution to the total magnetic scattering amplitude is of the order of a few percent at small scattering angles.

In principle, the contributions of the spin-other orbit and orbit-orbit terms of the atomic Hamiltonian can be included in the calculation of the relativistic corrections to the magnetic scattering amplitude. In light atoms the contribution of these terms is negligible in comparison to that of the kinetic-energy term. In heavy atoms, on the other hand, their contribution is smaller than that of the spin-orbit term.

C. Small-q approximation

If $q \ll 1/R$, where R is of the order of the atomic radius, only a few multipole moments contribute significantly to the magnetic scattering amplitude. We have seen that if the initial and final atomic states are of opposite parity the magnetic scattering amplitude, at small values of q, is determined by the transition matrix element of the electric dipole moment. This is the well-known dipole approximation in atomic spectroscopy. In fact, it can easily be seen [Eqs. (10), (9), (37)] that for $e^{i\vec{q}\cdot\vec{r}}\approx 1$ the operator $\vec{\mathcal{J}}_\perp$ is proportional to the transition matrix element of the electric dipole moment. For transitions within the l^n configuration the electric dipole contribution vanishes by parity and one must proceed to the next approximation $e^{i\vec{q}\cdot\vec{r}}\approx 1+i\vec{q}\cdot\vec{r}$. In this approximation only the magnetic dipole *and* electric quadrupole moments contribute to the scattering amplitude. In the present paper this approximation will be referred to as the small-q approximation to the magnetic scattering amplitude for transitions within the l^n electronic configuration.

For simplicity, we will adopt the extreme nonrelativistic approximation. The magnetic scattering amplitude in the small-q approximation will be given by Eq. (57) with

$$f^{\theta JM}_{\theta' J' M'}(\hat{q})=-i(|\gamma|r_0)\,\vec{\sigma}\cdot\sum_m\Big((\tfrac{8}{5}\pi)^{1/2}\vec{X}^*_{2m}(\hat{q})[R_2(2)(\theta JM|W_m^{(0,2)2}|\theta'J'M')+R_1(2,2)(\theta JM|W_m^{(1,2)2}|\theta'J'M')]$$
$$-(\tfrac{8}{3}\pi)^{1/2}[\hat{q}\times\mathbf{X}^*_{1m}(\hat{q})][R_0(1)\langle\theta JM|W_m^{(0,1)1}|\theta'J'M'\rangle+R_1(0,1)\langle\theta JM|W_m^{(1,0)1}|\theta'J'M'\rangle$$
$$+R_1(2,1)\langle\theta JM|W_m^{(1,2)1}|\theta'J'M'\rangle]\Big), \tag{66}$$

The first and second sums in this equation are the electric quadrupole and magnetic dipole contributions to the magnetic scattering amplitude, respectively. The radial matrix elements in Eq. (66) are obtained by using Eqs. (50)–(52):

$$
\begin{aligned}
R_0(1) &= \left(\frac{l(l+1)(2l+1)}{6}\right)^{1/2} (\bar{j}_0 + \bar{j}_2) \,, \\
R_1(0,1) &= \left(\frac{(2l+1)}{2}\right)^{1/2} \bar{j}_0 \,, \\
R_1(2,1) &= \frac{1}{2}\left(\frac{l(l+1)(2l+1)}{(2l-1)(2l+3)}\right)^{1/2} \bar{j}_2 \,, \\
R_2(2) &= i\left(\frac{l(l+1)(2l+1)}{120(2l-1)(2l+3)}\right)^{1/2} \bar{R}_2 \,.
\end{aligned}
\tag{67}
$$

It is seen that at small values of q the main contribution to the scattering amplitude arises from the first two terms of the magnetic dipole. A closed expression for the contribution of these terms can be obtained by exploiting the proportionality of the $W^{(0,1)1}$ and $W^{(1,0)1}$ tensors to the operators $\vec{L}$ and $\vec{S}$ of the atom:

$$
\begin{aligned}
W_m^{(0,1)1} &= \left(\frac{3}{2l(l+1)(2l+1)}\right)^{1/2} L_m \,, \\
W_m^{(1,0)1} &= \left(\frac{2}{2l+1}\right)^{1/2} S_m \,.
\end{aligned}
\tag{68}
$$

Using Eqs. (67), (68), and the Wigner-Eckart theorem, one obtains

$$
R_0(1)\langle \theta JM | W_m^{(0,1)1} | \theta' J' M' \rangle + R_1(0,1)\langle \theta JM | W_m^{(1,0)1} | \theta' J' M' \rangle = (-)^{J-M} \begin{pmatrix} J & 1 & J' \\ -M & m & M' \end{pmatrix} \tfrac{1}{2}(\theta J \| (\bar{j}_0 + \bar{j}_2)\vec{L} + \bar{j}_0(2\vec{S}) \| \theta' J') \,.
\tag{69}
$$

The reduced matrix elements in this equation can be expressed in terms of J, J', S, and L. In fact, if $J' = J \pm 1$,

$$
(\theta J \| \vec{L} \| \theta' J') = -(\theta J \| \vec{S} \| \theta' J') = \delta(\theta, \theta')(-)^{J-J_>+1}\left(\frac{(S+L+J_>+1)(L-S+J_>)(J_>+S-L)(S+L-J_>+1)}{4J_>}\right)^{1/2} \,,
\tag{70}
$$

where $J_>$ is the larger of J, J'. If $J = J'$, on the other hand,

$$
(\theta J \| \vec{L} \| \theta' J) = \delta(\theta, \theta') \frac{J(J+1) + L(L+1) - S(S+1)}{2J(J+1)} [J(J+1)(2J+1)]^{1/2}
\tag{71}
$$

and

$$
(\theta J \| \vec{S} \| \theta' J) = \delta(\theta, \theta') \frac{J(J+1) + S(S+1) - L(L+1)}{2J(J+1)} [J(J+1)(2J+1)]^{1/2} \,.
\tag{72}
$$

In any given problem the calculation of the magnetic scattering amplitude in the small-q approximation is reduced, by using Eq. (47), to evaluating the reduced matrix elements of the $W^{(0,2)2}$, $W^{(1,2)2}$, and $W^{(1,2)1}$ double tensors. These latter matrix elements can be obtained either from the existing tabulations or by using Eq. (55). It is seen that in the limit $q \to 0$ ($\bar{j}_0 \to 1$ and $\bar{j}_2 \to 0$), the magnetic scattering amplitude is proportional to $(\theta J \| \vec{L} + 2\vec{S} \| \theta J')$. As expected, the magnetic scattering amplitude in this limit is proportional to the magnetic moment of the atom.

The magnetic scattering amplitude [Eq. (66)] can be considerably simplified in the case of elastic scattering by an atom characterized by a single Russel-Saunders state $|\theta JM\rangle$. This is a good approximation for the elastic scattering of neutrons by rare-earth ions in their ground state. In this case the electric quadrupole contribution vanishes and only the $m = 0$ component of the magnetic dipole contributes to the scattering amplitude. The scattering amplitude can be written in a well-known form if one notes that

$$
\hat{q} \times \vec{X}^*_{10} = \frac{i\sqrt{3}}{2\sqrt{2\pi}} \vec{q}_m \,,
\tag{73}
$$

where

$$
\vec{q}_m = (\hat{q} \cdot \hat{\epsilon}_3)\hat{q} - \hat{\epsilon}_3
\tag{74}
$$

is the so-called magnetic scattering vector. Using Eqs. (69), (73), and (47) in Eq. (66), one obtains

$$f^{\theta JM}_{\theta JM}(\hat{q}) = -(|\gamma| r_0)(\vec{\sigma}\cdot\vec{q}_m)\frac{M}{[J(J+1)(2J+1)]^{1/2}}\left[\tfrac{1}{2}(\theta J||(\bar{j}_0+\bar{j}_2)\vec{L}+\bar{j}_0(2\vec{S})||\theta J)+(2J+1)\left(\frac{3l(l+1)(2l+1)}{4(2l-1)(2l+3)}\right)^{1/2} \times\bar{j}_2\begin{Bmatrix} S & S & 1\\ L & L & 2\\ J & J & 1\end{Bmatrix}(\theta||W^{(1,2)}||\theta)\right] . \tag{75}$$

It is seen that the magnetic scattering amplitude can be written in the conventional form $p(\vec{q})\vec{\sigma}\cdot\vec{q}_m$ where $p(\vec{q})$, normalized to 1 in the forward direction, defines the magnetic form factor $f_m(\vec{q})$. Using Eqs. (71) and (72) one obtains the following expression for the magnetic form factor:

$$f_m(\vec{q}) = \bar{j}_0 + \left[\frac{J(J+1)+L(L+1)-S(S+1)}{3J(J+1)+S(S+1)-L(L+1)} + \frac{2[J(J+1)(2J+1)]^{1/2}}{3J(J+1)+S(S+1)-L(L+1)}\left(\frac{3l(l+1)(2l+1)}{(2l-1)(2l+3)}\right)^{1/2}\begin{Bmatrix} S & S & 1\\ L & L & 2\\ J & J & 1\end{Bmatrix} \times(\theta||W^{(1,2)}||\theta)\right]\bar{j}_2 . \tag{76}$$

In the elastic scattering of neutrons by rare-earth ions in their ground state, this expression can be used to calculate the magnetic form factor at small scattering angles.

An important application of Eqs. (75) and (76) is in the calculation of the coherent amplitude for the elastic neutron scattering by an atom in an external field B. We will assume that, in the absence of the magnetic field, the atom is in a single Russel-Saunders state $|\theta JM\rangle$. This state, in the presence of the magnetic field, is split into $(2J+1)$ levels whose probability of occupation is proportional to $e^{-\beta M}$, where $\beta = g\mu_B B/kT$ and g is the Landé g factor of the atom. The coherent amplitude $\langle f^{\theta JM}_{\theta JM}(\vec{q})\rangle$ for the elastic neutron scattering by the atom is

$$\langle f^{\theta JM}_{\theta JM}(\vec{q})\rangle = \sum_M \frac{e^{-\beta M}}{(\sum_M e^{-\beta M})} f^{\theta JM}_{\theta JM}(\vec{q}) , \tag{77}$$

where $f^{\theta JM}_{\theta JM}(\vec{q})$ is given by Eq. (59).

This expression can be simplified at high temperatures, $\beta \ll 1$. In this case $e^{-\beta M} \simeq 1-\beta M$, and it can be seen that only the magnetic dipole term ($k=1$) in Eq. (59) contributes to the coherent scattering amplitude. Thus, for $\beta \ll 1$ the coherent scattering amplitude is inversely proportional to the temperature and its angular dependence is given by Eq. (76). Note that this result is not restricted to small scattering vectors.

In this section we illustrated the formalism by examining some simple examples. However, the advantages of this formalism over more conventional techniques becomes evident when one examines the magnetic scattering of neutrons by complex atoms.

IV. SUMMARY

We showed that the magnetic scattering of neutrons by an arbitrary system of particles can be investigated by techniques similar to those used in studying the interaction of radiation with the system. It has been shown, in fact, that the magnetic scattering amplitude can be expressed [Eq. (24)] in terms of the multipole moments of the system. This expression for the magnetic scattering amplitude exhibits explicitly the main physical features of the magnetic scattering of neutrons. The magnetic scattering amplitude is the scalar product of the magnetic moment of the neutron with a vector field *transverse* to the scattering vector. The separation of the magnetic scattering amplitude into a spin magnetization and a convection current contribution arises naturally as a result of the same separation in the multipole moments. The number of multipoles that contribute to the magnetic scattering amplitude is limited by the symmetry properties of the states of the system, in particular, parity and angular momentum conservation [Eqs. (25), (26)]. The calculation of the magnetic scattering amplitude is thus reduced to evaluating matrix elements, between the initial and final states of the scattering system, of a limited number of multipole moments. In any given problem the evaluation of these matrix elements is a straightforward application of well-known techniques in modern spectroscopy. In the small-q approximation the scattering amplitude is determined by the transition matrix element of the electric dipole moment if the parity of the scattering system changes in the scattering process. If the parity of the system, on the other hand, does not change during the scattering process, the magnetic scattering amplitude in the small-q approximation is determined by the transition matrix element of the magnetic dipole moment.

The formalism has been applied to the magnetic scattering of neutrons by an atom in the l^n electronic configuration. If the spin-other-orbit and orbit-orbit interactions in the atomic Hamiltonian can be neglected only even-order electric and odd-order magnetic multipoles, whose order of multipolarity is less than or equal to $2l+1$, contribute to the magnetic scattering amplitude. The magnetic scattering amplitude, in this case, can be expressed in terms of the $W^{(0,k)}$ and $W^{(1,k')}$ (k' even) Racah double tensors. The calculation of the matrix elements of these tensors is considerably simplified by the selection rules based on the groups Sp($4l+2$), R($2l+1$), R(3), and in the case of f electrons the special group G_2. The nonvanishing matrix elements needed for the evaluation of the magnetic scattering amplitude are then calculated [Eq. (55)] or they are obtained directly from the tabulations. The corrections to the magnetic scattering amplitude due to the spin-orbit and mass correction terms of the Hamiltonian have been calculated. These effects can be of importance in the investigation of the conduction electron polarization effects in heavy metals.

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APPENDIX

In this appendix we define the Racah tensors $W^{(k',k'')}$ and $W^{(k',k'')k}$. Following Judd[14] we introduce the unit tensors $t^{k'}$ and $v^{k''}$ which act in the spin and orbital spaces, respectively, of a single electron, and which are defined by their reduced matrix elements

$$(s\,||t^{k'}||\,s')=(2k'+1)^{1/2}\delta(s,s')\;,$$

$$(l\,||v^{k''}||\,l')=(2k''+1)^{1/2}\delta(l,l')\;.$$

The $(2k'+1)(2k''+1)$ products

$$w^{(k',k'')}_{m'm''}=t^{k'}_{m'}v^{k''}_{m''}\quad(-k'\le m'\le k';\;-k''\le m''\le k'') \tag{A1}$$

define the components of the double tensor $w^{(k',k'')}$ whose reduced matrix elements are

$$(sl\,||w^{(k',k'')}||\,s'l')=[k',k'']^{1/2}\delta(l,l')\delta(s,s')\;.$$

The tensor $w^{(k',k'')k}$ is defined as the tensor product of the tensors $t^{k'}$ and $v^{k''}$

$$w^{(k',k'')k}_{m}=[t^{k'}\times v^{k''}]^{k}_{m} \tag{A2}$$

Using the single-particle tensors defined by (A1) and (A2) we can define the one-particle operators $W^{(k',k'')}$ and $W^{(k',k'')k}$ by

$$W^{(k',k'')}_{m'm''}=\sum_i w^{(k',k'')}_{m'm''}(i)\;, \tag{A3}$$

$$W^{(k',k'')k}_{m}=\sum_i w^{(k',k'')k}_{m}(i)\;, \tag{A4}$$

where the sum is over the electrons of the l^n configuration.

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