Polarization dependence of magnetic x-ray scattering

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(Received 12 June 1987)

We calculate the polarization dependence of the x-ray scattering cross section, including magnetic terms, using the Poincaré representation for the polarization. General expressions are given for the polarization dependence of the cross section of the pure magnetic scattering and of the interference between charge and magnetic scattering, and for the polarization of the scattered beam in both cases. These expressions are compared to the equivalent results for magnetic neutron scattering. The general results are then specialized to several typical cases, including the scattering of linearly and circularly polarized radiation from spiral, uniaxially modulated, and ferromagnetic structures. It is shown that detailed magnetic-structure determinations are possible using synchrotron radiation. It is further demonstrated that the orbital- and spin-angular-momentum contributions of both ferromagnets and antiferromagnets may be separately measured in a variety of simple geometries. It is found that, although the efficiency is very low, linearly polarized radiation can be completely converted to circular polarization by scattering from a magnetic spiral. Finally, it is shown that, in addition to the interference between charge and magnetic scattering, there is an interference involving the spin- and orbital-angular-momentum scattering, which can couple moments in different spatial directions.

I. INTRODUCTION

In the last several years magnetic x-ray experiments using synchrotron radiation have been performed on a steadily growing number of magnetic systems. These experiments have included high-resolution studies of the pure magnetic scattering in antiferromagnets[1-3] as well as of the interference between charge and magnetic scattering in bulk and thin-film ferromagnets.[4-6] Spin-dependent Compton- (Refs. 7 and 8) and resonance-magnetic-scattering[9] studies have been performed on a variety of ferromagnets. The high brightness of synchrotron radiation sources has been an important factor in the success of many of these experiments. It has now become apparent that the polarization dependence of the magnetic cross section can also be exploited in even more detailed studies. First, the use of the polarization dependence provides a natural technique for determining magnetic structures by x-ray scattering. Beyond this, there are novel possibilities which arise from the well-defined polarization characteristics of synchrotron radiation. For example, using the high degree of linear polarization of the incident beam it has been possible in synchrotron experiments to distinguish between charge peaks, arising from lattice modulations, and magnetic peaks, in a spiral magnetic structure.[2,10] This distinction was crucial to the interpretation of the diffraction pattern in recent studies of rare-earth metals. Furthermore, it has been suggested that by analyzing the polarization of the scattered beam, it should be possible to separately measure the spin and orbital contributions to the cross section.[11] This separation is not directly possible by neutron-scattering techniques and is important to a fundamental understanding of the electronic properties of magnetic materials. Along these lines, we note the elegant experiments of Brunel *et al.*[5] in which the scattering of circularly polarized synchrotron radiation was explicitly observed in a powdered ferrite.

In this paper we calculate the polarization dependence of the x-ray scattering cross section, including magnetic terms, using the Poincaré representation for the polarization. General expressions are given for the polarization dependence of the cross section of the pure magnetic scattering and of the interference between charge and magnetic scattering, and for the polarization of the scattered beam in both cases. These expressions are compared to the equivalent results for magnetic neutron scattering. The general results are then specialized to several typical cases, including the scattering of linearly and circularly polarized radiation from spiral, uniaxially modulated, and ferromagnetic structures. It is shown that detailed magnetic-structure determinations are possible using synchrotron radiation by measuring the polarization dependence of the magnetic and interference cross sections, and by analyzing the polarization of the magnetically scattered beam. It is further demonstrated that the orbital- and spin-angular-momentum contributions of both ferromagnets and antiferromagnets may be separately measured in a variety of simple geometries. It is found that, although the efficiency is very low ($<\sim 10^{-6}$), linearly polarized radiation can be completely converted to circular polarization by scattering from a magnetic spiral. Finally, we note that, in addition to the interference between charge and magnetic scattering, there is an interference involving the spin- and orbital-angular-momentum scattering which can couple moments in different spatial directions.

Although particular features of the polarization dependence of the cross section have already appeared in earlier papers,[12,1,4,5,11] the general case, explicitly including the orbital angular momentum and the final polarization, has not been previously published. The present results are surprisingly simple and will be important for their utility in suggesting and in analyzing the results of synchrotron experiments. The general results for the x-ray cross section are summarized in Eqs. (6), (7), and (9). This expression is expanded using the Poincaré representation in Eqs. (11), (12), (14), and (16). General expressions for the final polarization of the magnetic scattering are given in Eq. (13).

II. CROSS SECTION

The cross section for magnetic scattering of photons by free charges and atoms has been discussed by a number of authors.[13,14,12,4,5,11] In the following we reproduce the expression of Blume[11] obtained by a nonrelativistic calculation of the cross section using perturbation theory. In the limit of high photon energy the cross section for elastic scattering is

$$\left.\frac{d^2\sigma}{d\Omega' dE'}\right|_{\lambda\to\lambda',a\to b} = \left[\frac{e^2}{mc^2}\right]^2 \left| \left\langle b \left| \sum_j e^{i\mathbf{K}\cdot\mathbf{r}_j} \right| a\right\rangle \hat{\epsilon}\cdot\hat{\epsilon}' - \frac{i\hbar\omega}{mc^2}\left\langle b\left|\sum_j e^{i\mathbf{K}\cdot\mathbf{r}_j}\left[\frac{i\mathbf{K}\times\mathbf{p}_j}{\hbar k^2}\cdot\mathbf{A}+\mathbf{s}_j\cdot\mathbf{B}\right]\right|a\right\rangle\right|^2 \delta(E_a-E_b-(\hbar\omega'_k-\hbar\omega_k)) \;, \qquad (1)$$

where $\mathbf{A}=\hat{\epsilon}'\times\hat{\epsilon}$ and

$$\mathbf{B}=\hat{\epsilon}'\times\hat{\epsilon}+(\hat{\mathbf{k}}'\times\hat{\epsilon}')(\hat{\mathbf{k}}'\cdot\hat{\epsilon})-(\hat{\mathbf{k}}\times\hat{\epsilon})(\hat{\mathbf{k}}\cdot\hat{\epsilon}') -(\hat{\mathbf{k}}'\times\hat{\epsilon}')\times(\hat{\mathbf{k}}\times\hat{\epsilon}) \;.$$

Here the sum is taken over all electrons j, $\mathbf{K}=\mathbf{k}-\mathbf{k}'$ is the momentum transfer, $\hbar\omega$ ($\hbar\omega'$) is the incident (scattered) photon energy, a (b) is the initial (final) state of the scatterer, $\hat{\epsilon}$ ($\hat{\epsilon}'$) is the initial (scattered) polarization, and $\mathbf{p}_j$ is the electronic momentum. The geometry and convections used here are illustrated in Fig. 1. The modulus of the first term on the right-hand side of the equation gives the usual Thomson cross section for charge scattering and depends on the Fourier transform of the charge density. The modulus of the second term, which is reduced from the first by $(\hbar\omega/mc^2)^2$, describes the pure magnetic scattering and depends on the Fourier transforms of the spin and orbital magnetization densities. In addition, there is an interference term proportional to $(i\hbar\omega/cm^2)$, involving the products of charge and magnetic densities. We first develop an expression for the orbital momentum.

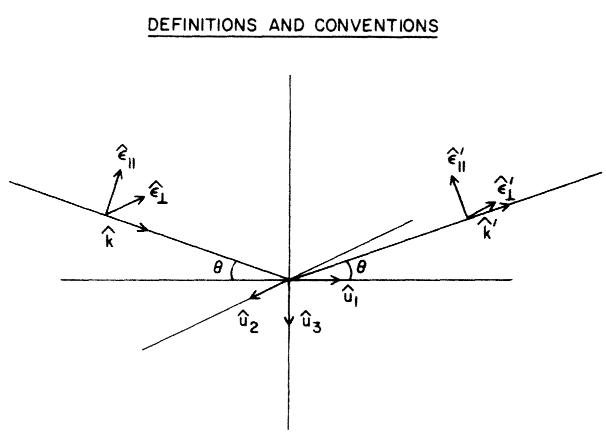

FIG. 1. The definitions and conventions used in this paper. $\hat{\mathbf{k}}$ and $\hat{\mathbf{k}}'$ are the incident and scattered wavevectors and 2θ is the scattering angle. $\hat{\epsilon}_{\perp}$ and $\hat{\epsilon}_{\parallel}$ are the components of the polarization perpendicular and parallel to the diffraction plane (spanned by $\hat{\mathbf{k}}$ and $\hat{\mathbf{k}}'$). The $\hat{\mathbf{U}}_i$'s define a basis for the magnetic structure which is expressed in terms of the incident and scattered wavevectors: $\hat{\mathbf{U}}_1=(\hat{\mathbf{k}}+\hat{\mathbf{k}}')/2\cos\theta$, $\hat{\mathbf{U}}_2=\hat{\mathbf{k}}\times\hat{\mathbf{k}}'/\sin 2\theta$, $\hat{\mathbf{U}}_3=(\hat{\mathbf{k}}-\hat{\mathbf{k}}')/2\sin\theta$. By these conventions we also have $\hat{\epsilon}_{\perp}=-\hat{\mathbf{U}}_2$, $\hat{\epsilon}'_{\perp}=-\hat{\mathbf{U}}_2$, $\hat{\epsilon}_{\parallel}=\sin\theta\,\hat{\mathbf{U}}_1-\cos\theta\,\hat{\mathbf{U}}_3$, and $\hat{\epsilon}'_{\parallel}=-(\sin\theta\,\hat{\mathbf{U}}_1+\cos\theta\,\hat{\mathbf{U}}_3)$.

Rewriting the orbital term:

$$\sum_j e^{i\mathbf{K}\cdot\mathbf{r}_j}\frac{i(\mathbf{K}\times\mathbf{p}_j)}{k^2}\cdot\mathbf{A}\to\sum_j e^{i\mathbf{K}\cdot\mathbf{r}_j}\left[\frac{-i}{K}\hat{\mathbf{K}}\times\mathbf{p}_j\right]\cdot\mathbf{A}' \;,$$

where $\mathbf{A}'=-(K^2/k^2)\mathbf{A}=-4(\sin^2\theta)(\hat{\epsilon}'\times\hat{\epsilon})$ and 2θ is the scattering angle. This expression is analogous to that encountered in neutron scattering and for elastic scattering may be rewritten as[15–18]

$$-\left\langle a\left|\sum_j e^{i\mathbf{K}\cdot\mathbf{r}_j}\frac{(i\hat{\mathbf{K}}\times\mathbf{p}_j)}{K}\right|a\right\rangle\to\tfrac{1}{2}\hat{\mathbf{K}}\times[\mathbf{L}(\mathbf{K})\times\hat{\mathbf{K}}] \;,$$

where $\mathbf{L}(\mathbf{K})$ is the Fourier transform of the atomic-orbital magnetization density.[15–17] Explicitly,[15]

$$\mathbf{L}(\mathbf{K})=\frac{1}{2}\langle a \mid \sum_j [f(\mathbf{K}\cdot\mathbf{r}_j)\mathbf{l}_j+\mathbf{l}_j f(\mathbf{K}\cdot\mathbf{r}_j)] \mid a\rangle \;,$$

where

$$f(x)=2\sum_{n=0}^{\infty}\frac{(ix)^n}{(n+2)n!} \;.$$

As a consequence of the vector product the contribution of the orbital term in the direction of the momentum transfer $\mathbf{K}$ is zero, just as with neutron scattering. Through the use of simple vector identities,

$$\hat{\mathbf{K}}\times(\mathbf{L}\times\hat{\mathbf{K}})\cdot\mathbf{A}'=\mathbf{L}\cdot[\mathbf{A}'-(\mathbf{A}\cdot\mathbf{K})\hat{\mathbf{K}}]\equiv\mathbf{L}\cdot\mathbf{A}'' \qquad (2)$$

with

$$\mathbf{A}''=\mathbf{A}'-(\mathbf{A}'\cdot\hat{\mathbf{K}})\hat{\mathbf{K}}$$
$$=2(1-\hat{\mathbf{k}}\cdot\hat{\mathbf{k}}')(\hat{\boldsymbol{\epsilon}}'\times\hat{\boldsymbol{\epsilon}})$$
$$-(\hat{\mathbf{k}}\times\hat{\boldsymbol{\epsilon}})(\hat{\mathbf{k}}\cdot\hat{\boldsymbol{\epsilon}}')+(\hat{\mathbf{k}}'\times\hat{\boldsymbol{\epsilon}}')(\hat{\mathbf{k}}'\cdot\hat{\boldsymbol{\epsilon}})\ .$$

Defining the Fourier transform of the spin density,

$$\mathbf{S}(\mathbf{K})=\left\langle a\left|\sum_j e^{i\mathbf{K}\cdot\mathbf{r}_j}\mathbf{s}_j\right|a\right\rangle\ ,$$

the magnetization-dependent part $\langle M_m\rangle$ of the x-ray cross section may be written explicitly in terms of $\mathbf{L}(\mathbf{K})$ and $\mathbf{S}(\mathbf{K})$:

$$\langle M_m\rangle=\tfrac{1}{2}\mathbf{L}(\mathbf{K})\cdot\mathbf{A}''+\mathbf{S}(\mathbf{K})\cdot\mathbf{B}\ . \tag{3}$$

$\mathbf{A}''$ and $\mathbf{B}$ are given in Eqs. (1) and (2). It is clear that the orbital and spin contributions to the x-ray cross section are different and so they may be distinguished by analyzing the polarization of the scattered beam. This difference does not appear in neutron scattering, where the interaction is purely magnetic in origin. Explicitly, the expression for neutron magnetic scattering is[11]

$$\langle M_n\rangle=\hat{\mathbf{K}}\times\{[\tfrac{1}{2}\mathbf{L}(\mathbf{K})+\mathbf{S}(\mathbf{K})]\times\hat{\mathbf{K}}\}\cdot\boldsymbol{\sigma}$$
$$\equiv[\tfrac{1}{2}\mathbf{L}(\mathbf{K})+\mathbf{S}(\mathbf{K})]\cdot\mathbf{C}\ ,$$

where $\boldsymbol{\sigma}$ is the neutron spin operator and $\mathbf{C}=[\hat{\mathbf{K}}\times(\boldsymbol{\sigma}\times\hat{\mathbf{K}})]$. It is seen that the polarization dependence of the spin magnetization density is identical to that for the orbital magnetization density. The difference in the cross sections for x-ray scattering and neutron scattering arises because the x-ray interacts both with the charge (through the electric field and its gradients) and with the magnetic moment (through the magnetic fields and their gradients). Thus, the Lorentz force, for example, affects only the orbital magnetic moment and not the spin, to first order in $(\hbar\omega/mc^2)$.

From the point of view of performing synchrotron experiments it is convenient to express the vectors $\mathbf{A}''$ and $\mathbf{B}$ as 2×2 matrices in a basis whose components are parallel and perpendicular to the diffraction plane (see Fig. 1). Then

$$\mathbf{A}''=\begin{bmatrix}A''_{\perp\perp} & A''_{\perp\parallel}\\ A''_{\parallel\perp} & A''_{\parallel\parallel}\end{bmatrix}=\frac{K^2}{2k^2}\begin{bmatrix}0 & -(\hat{\mathbf{k}}+\hat{\mathbf{k}}')\\ \hat{\mathbf{k}}+\hat{\mathbf{k}}' & 2\hat{\mathbf{k}}\times\hat{\mathbf{k}}'\end{bmatrix}\ , \tag{4a}$$

$$\mathbf{B}=\begin{bmatrix}B_{\perp\perp} & B_{\perp\parallel}\\ B_{\parallel\perp} & B_{\parallel\parallel}\end{bmatrix}=\begin{bmatrix}\hat{\mathbf{k}}\times\hat{\mathbf{k}}' & -\hat{\mathbf{k}}'(1-\hat{\mathbf{k}}\cdot\hat{\mathbf{k}}')\\ \hat{\mathbf{k}}(1-\hat{\mathbf{k}}\cdot\hat{\mathbf{k}}') & \hat{\mathbf{k}}\times\hat{\mathbf{k}}'\end{bmatrix}\ . \tag{4b}$$

This representation of the matrix $\mathbf{B}$ has been given by de Bergevin and Brunel.[1,4] $\langle M_m\rangle$ may now be written as

$$\langle M_m\rangle=\begin{bmatrix}\langle M_m\rangle_{\perp\perp} & \langle M_m\rangle_{\perp\parallel}\\ \langle M_m\rangle_{\parallel\perp} & \langle M_m\rangle_{\parallel\perp}\end{bmatrix}=\begin{bmatrix}\mathbf{S}\cdot(\hat{\mathbf{k}}\times\hat{\mathbf{k}}') & -\dfrac{K^2}{2k^2}\left[\left[\dfrac{\mathbf{L}(\mathbf{K})}{2}+\mathbf{S}(\mathbf{K})\right]\cdot\hat{\mathbf{k}}'+\dfrac{\mathbf{L}(\mathbf{K})}{2}\cdot\hat{\mathbf{k}}\right]\\ \dfrac{K^2}{2k^2}\left[\left[\dfrac{\mathrm{L}}{2}\mathbf{L}(\mathbf{K})+\mathbf{S}(\mathbf{K})\right]\cdot\hat{\mathbf{k}}+\dfrac{\mathbf{L}(\mathbf{K})}{2}\cdot\hat{\mathbf{k}}'\right] & \left[\dfrac{K^2}{2k^2}\mathbf{L}(\mathbf{K})+\mathbf{S}(\mathbf{K})\right]\cdot(\hat{\mathbf{k}}\times\hat{\mathbf{k}}')\end{bmatrix}. \tag{5}$$

The diagonal matrix element involve magnetization density oriented only in the direction perpendicular to the diffraction plane, while the off-diagonal matrix elements involve magnetization density oriented only within the diffraction plane. Further, $\langle M_m\rangle_{\perp\perp}$ is independent of $\mathbf{L}(\mathbf{K})$. For general magnetic structures this same expression holds with $\mathbf{L}(\mathbf{K})$ and $\mathbf{S}(\mathbf{K})$ representing the complex structure factors. Expressing the component of the magnetic structure in the basis defined in Fig. 1 we have

$$\langle M_m\rangle=\begin{bmatrix}(\sin2\theta)S_2 & -2(\sin^2\theta)[(\cos\theta)(L_1+S_1)-(\sin\theta)S_3]\\ 2(\sin^2\theta)[(\cos\theta)(L_1+S_1)+(\sin\theta)S_3] & (\sin2\theta)[2(\sin^2\theta)L_2+S_2]\end{bmatrix}\ , \tag{6}$$

where 2θ is the scattering angle and we have used $\frac{1}{2}(K/k)^2=2\sin^2\theta$. In this basis we may also write the interaction matrix describing the charge scattering:

$$\langle M_c\rangle=\rho(\mathbf{K})\begin{bmatrix}1 & 0\\ 0 & \cos2\theta\end{bmatrix}\ , \tag{7}$$

where $\rho(\mathbf{K})$ is the Fourier transform of the electronic charge density.

III. POINCARÉ REPRESENTATION

Having general expressions for the matrices $\langle M_m\rangle$ and $\langle M_c\rangle$, it is now possible to calculate the cross section and the final polarization for arbitrary incident polarization and for any magnetic structure. It is convenient to introduce the Poincaré representation for the polarization and the density matrix for the incident beam. The Poincaré representation is particularly useful as it applies to both completely and partially polarized incident radiation and involves only variables which are measured in experiments. A general discussion of these techniques has been given by Fano.[18] A discussion of their application to neutron scattering and to the transmission of x-rays through matter has been given by Blume and Kistner.[17] The first application of the Poincaré representation to magnetic x-ray scattering was by de Bergevin and Brunel.[1]

We write the expression for elastic scattering by explicitly introducing the initial λ and final λ' polarizations and taking the expectation value of M in the initial and final state $|a\rangle$ of the scatterer:

$$\frac{d\sigma}{d\Omega'}=\left[\frac{e^2}{mc^2}\right]^2\sum_{\lambda,\lambda'}p_\lambda\left|\langle\lambda'|\langle M_c\rangle|\lambda\rangle-\frac{i\hbar\omega}{mc^2}\langle\lambda'|\langle M_m\rangle|\lambda\rangle\right|^2, \tag{8}$$

where

$$\langle\lambda'|\langle M_c\rangle|\lambda\rangle=\left\langle a\left|\sum e^{i\mathbf{K}\cdot\mathbf{r}_j}\right|a\right\rangle\hat{\epsilon}_\lambda\cdot\hat{\epsilon}_{\lambda'}$$

and

$$\langle\lambda'|\langle M_m\rangle|\lambda\rangle=\tfrac{1}{2}\mathbf{L}(\mathbf{K})\cdot\mathbf{A}''_{\lambda'\lambda}+\mathbf{S}(\mathbf{K})\cdot\mathbf{B}_{\lambda'\lambda}\,.$$

Here p_λ is the probability for incident polarization λ. We next define the (2×2) density matrix ρ for the incident beam by

$$\rho=\sum_\lambda|\lambda\rangle p_\lambda\langle\lambda|\,.$$

Evaluating, we obtain the general expression for the equilibrium differential cross section for elastic scattering:

$$\frac{d\sigma}{d\Omega}=\left[\frac{e^2}{mc^2}\right]^2\operatorname{tr}\left[\left\langle M_c-\frac{i\hbar\omega}{mc^2}M_m\right\rangle\rho\left\langle M_c-\frac{i\hbar\omega}{mc^2}M_m\right\rangle^\dagger\right]$$
$$=\left[\frac{e}{mc^2}\right]^2\operatorname{tr}\left[\langle M_c\rangle\rho\langle M_c^\dagger\rangle-\frac{i\hbar\omega}{mc^2}(\langle M_m\rangle\rho\langle M_c^\dagger\rangle-\langle M_c\rangle\rho\langle M_m^\dagger\rangle)+\left[\frac{\hbar\omega}{mc^2}\right]^2\langle M_m\rangle\rho\langle M_m^\dagger\rangle\right]. \tag{9}$$

$\langle M^\dagger\rangle$ is the Hermitian conjugate of $\langle M\rangle$.

Following Fano,[18] we now develop the expression for the density matrix. Because the density matrix is the averaged outer product of a two-dimensional vector, it is Hermitian, and consequently may be expressed in terms of the unit matrix and the Pauli matrices:

$$\rho=\tfrac{1}{2}(1+\mathbf{P}\cdot\boldsymbol{\sigma})\ \text{ where }\begin{cases}\mathbf{P}=(P_\xi,P_\eta,P_\zeta),\\ \boldsymbol{\sigma}=(\sigma_\xi,\sigma_\eta,\sigma_\zeta).\end{cases}$$

Here $\boldsymbol{\sigma}$ represents the Pauli matrices, I_0 is the total intensity, and P_ξ, P_η, and P_ζ give the Poincaré-Stokes representation of the polarization. We write the components of the Poincaré vector $\mathbf{P}$ using Greek symbols to emphasize that it is not a vector in real space. In the usual Cartesian coordinate system we have

$$\rho=\frac{1}{2}\begin{bmatrix}1+P_\zeta & P_\xi-iP_\eta\\ P_\xi+iP_\eta & 1-P_\zeta\end{bmatrix},$$

where $\sigma_\xi=\left(\begin{smallmatrix}0&1\\1&0\end{smallmatrix}\right)$, $\sigma_\eta=\left(\begin{smallmatrix}0&-i\\i&0\end{smallmatrix}\right)$, and $\sigma_\zeta=\left(\begin{smallmatrix}1&0\\0&-1\end{smallmatrix}\right)$. Taking $\hat{\epsilon}_\perp$ and $\hat{\epsilon}_\parallel$ as two orthogonal unit vectors perpendicular to the beam direction defined in Fig. 1 (with $\mathbf{E}=E_1\hat{\epsilon}_\perp+E_2\hat{\epsilon}_\parallel$), the components of $\mathbf{P}$ are defined as follows. Let I_1 be the difference between the light intensity with linear polarization parallel to a vector oriented 45° to $\hat{\epsilon}_\perp$ and the intensity with linear polarization parallel to a vector oriented 45° to $\hat{\epsilon}_\parallel$. Then

$$P_\xi=\frac{I_1}{I_0}=\frac{|E_1+E_2|^2-|E_1-E_2|^2}{4(E_1|^2+|E_2|^2)}=\frac{\operatorname{Re}(E_1^*E_2)}{|E_1|^2+|E_2|^2}\,.$$

Let I_2 be the difference between the light intensity with left circular polarization and the intensity with right circular polarization. Then

$$P_\eta=\frac{I_2}{I_0}=\frac{|E_1+iE_2|^2-|E_1-iE_2|^2}{4(|E_1|^2+|E_2|^2)}=\frac{\operatorname{Im}(E_1^*E_2)}{|E_1|^2+|E_2|^2}\,.$$

Let I_3 be the difference between the light intensity with linear polarization parallel to $\hat{\epsilon}_\perp$ and the intensity with polarization parallel to $\hat{\epsilon}_\parallel$. Then

$$P_\zeta=\frac{I_3}{I_0}=\frac{|E_1|^2-|E_2|^2}{|E_1|^2+|E_2|^2}\,.$$

It follows that $P_\xi=+1$ (-1) represent linear polarization at angles $+45°$ $(-45°)$ to the $\hat{\mathbf{U}}_3$ axis, $P_\eta=+1$ (-1) represent left (right) circular polarization, and $P_\zeta=+1$ (-1) represent linear polarization along the $\hat{\mathbf{U}}_2$ $(\hat{\mathbf{U}}_3)$ axis, respectively (see Fig. 1). If $|\mathbf{P}|=1$, the beam is completely polarized; if $|\mathbf{P}|\le1$, the beam is partially polarized; and if $|\mathbf{P}|=0$, the beam is unpolarized. The vector $\mathbf{P}$ may simply be thought of as a vector in an abstract space which is rotated upon scattering, as shown in Fig. 2.

Once the Poincaré vector $\mathbf{P}$ and the total intensity I_0 are specified, the polarization is completely characterized in terms of measurable intensities. To calculate the density matrix and final polarization after scattering we will require that[18]

$$\mathbf{P}=\operatorname{tr}(\sigma\rho)\,, \tag{10a}$$

$$\rho'=M\rho M^\dagger\,, \tag{10b}$$

$$\frac{d\sigma}{d\Omega}=\left[\frac{e^2}{mc^2}\right]^2\operatorname{tr}(\rho')\,, \tag{10c}$$

$$\mathbf{P}'=\frac{\operatorname{tr}(\sigma\rho')}{\operatorname{tr}(\rho')}\,. \tag{10d}$$

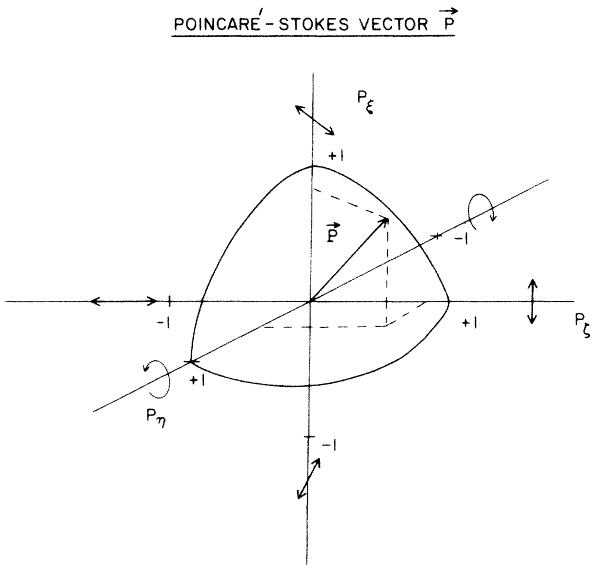

FIG. 2. The Poincaré sphere. The polarization is completely characterized by the values of the Poincaré coefficients P_ξ, P_η, P_ζ, and by the total intensity I_0. By definition, $|\mathbf{P}| \le 1$.

IV. RESULTS

In this section we calculate the general expressions for the differential cross section and the final polarization for each of the terms in Eq. (8), assuming arbitrary incident polarization $\mathbf{P}=(P_\xi,P_\eta,P_\zeta)$. For the purpose of discussion, the terms multiplying P_ξ will be referred to as the 45°-linear component, the terms multiplying P_η will be referred to as the circular component, and the terms multiplying P_ζ will be referred to as the linear component. The terms not associated with a Poincaré coefficient are referred to as the unpolarized component.

A. Charge scattering

This case is straightforward and gives the expected results. From Eq. (7) and (10) we write

$$\frac{d\sigma}{d\Omega}=\left[\frac{e^2}{mc^2}\right]^2 \mathrm{tr}\langle M_c\rangle\rho\langle M_c^\dagger\rangle$$

$$=\left[\frac{e^2}{mc^2}\right]^2 \tfrac{1}{2}|\rho(\mathbf{K})|^2[1+\cos^2 2\theta+P_\zeta(1-\cos^2 2\theta)]\,, \tag{11}$$

independent of P_ξ and P_η. The components of the final polarization $\mathbf{P}'$ may be found using Eq. (10d):

$$P'_\xi=\frac{2(\cos 2\theta)P_\xi}{1+\cos^2 2\theta+P_\zeta(1-\cos^2 2\theta)}\,,$$

$$P'_\eta=\frac{2(\cos 2\theta)P_\eta}{1+\cos^2 2\theta+P_\zeta(1-\cos^2 2\theta)}\,,$$

$$P'_\zeta=\frac{1-\cos^2 2\theta+P_\zeta(1+\cos^2 2\theta)}{1+\cos^2 2\theta+P_\zeta(1-\cos^2 2\theta)}\,.$$

B. Pure magnetic scattering

From Eq. (9) we write

$$\frac{d\sigma}{d\Omega}=\left[\frac{e^2}{mc^2}\right]^2\left[\frac{\hbar\omega}{mc^2}\right]^2 \mathrm{tr}\{\langle M_m\rangle\rho\langle M_m^\dagger\rangle\}$$

$$\to\left[\frac{e^2}{mc^2}\right]^2\left[\frac{\hbar\omega}{mc^2}\right]^2 \tfrac{1}{2}\{(1+P_\zeta)(|m_{11}|^2+|m_{21}|^2)+(1-P_\zeta)(|m_{12}|^2+|m_{22}|^2)$$

$$+2\,\mathrm{Re}[(P_\xi+iP_\eta)(m_{11}^*m_{12}+m_{21}^*m_{22})]\}\,, \tag{12}$$

where we have left the result in terms of $m_{i,j}$, the elements of $\langle M_m\rangle$. This form permits several general conclusions to be drawn below, and makes writing the cross section for magnetic structures and orientations not considered in the examples straightforward. Similarly, we expand Eq. (10d) to obtain

$$\frac{d\sigma}{d\Omega}P'_\xi=\left[\frac{e^2}{mc^2}\right]^2\left[\frac{\hbar\omega}{mc^2}\right]^2\{(1+P_\zeta)\mathrm{Re}(m_{11}^*m_{21})+(1-P_\zeta)\mathrm{Re}(m_{12}^*m_{22})+\mathrm{Re}[(P_\xi+iP_\eta)(m_{11}^*m_{22}+m_{21}^*m_{12})]\}\,,$$

$$\frac{d\sigma}{d\Omega}P'_\eta=\left[\frac{e^2}{mc^2}\right]^2\left[\frac{\hbar\omega}{mc^2}\right]^2\{(1+P_\zeta)\mathrm{Im}(m_{11}^*m_{21})+(1-P_\zeta)\mathrm{Im}(m_{12}^*m_{22})+\mathrm{Im}[(P_\xi+iP_\eta)(m_{11}^*m_{22}-m_{21}^*m_{12})]\}\,, \tag{13}$$

$$\frac{d\sigma}{d\Omega}P'_\zeta=\frac{1}{2}\left[\frac{e^2}{mc^2}\right]^2\left[\frac{\hbar\omega}{mc^2}\right]^2\{(1+P_\zeta)(|m_{11}|^2-|m_{21}|^2)+(1-P_\zeta)(|m_{12}|^2-|m_{22}|^2)$$

$$+2\,\mathrm{Re}[(P_\xi+iP_\eta)(m_{11}^*m_{12}-m_{21}^*m_{22})]\}\,.$$

In each of the expressions above the linear and unpolarized components and the circular and 45°-linear components display a simple symmetry, involving the same products of matrix elements. If $\langle M_m \rangle$ is diagonal, then there are no contributions to the cross section from the 45°-linear or the circular components. If $\langle M_m \rangle$ is nondiagonal, there are again no contributions from the 45°-linear or circular components. Similarly, if $\langle M_m \rangle$ is diagonal or nondiagonal and $P_\xi = P_\eta = 0$ then $P'_\xi = P'_\eta = 0$.

A feature of the cross section which is special to magnetic x-ray scattering is the existence of an interference between the spin and orbital angular momentum, which may couple moments in different spatial directions. These terms arise in each of the matrix element products above, except $|m_{11}|^2$. To illustrate, we write the general expression for the magnetic cross section by substituting Eq. (6) for $\langle M_m \rangle$ in Eq. (12):

$$\begin{aligned}\frac{d\sigma}{d\Omega} = \frac{1}{2}\left[\frac{e^2}{mc^2}\right]^2\left[\frac{\hbar\omega}{mc^2}\right]^2 &[(1+P_\xi)\{(\sin^2 2\theta)[\,|S_2|^2+(\sin^2\theta)|L_1+S_1|^2]+4(\sin^6\theta)|S_3|^2 \\ &+4(\sin 2\theta)(\sin^4\theta)[(L'_1+S'_1)S'_3+(L''_1+S''_1)S''_3]\} \\ &+(1-P_\xi)\{(\sin^2 2\theta)[\,|2(\sin^2\theta)L_2+S_2|^2+(\sin^2\theta)|L_1+S_1|^2]+4(\sin^6\theta)|S_3|^2 \\ &-4(\sin 2\theta)(\sin^4\theta)[(L'_1+S'_1)S'_3+(L''_1+S''_1)S''_3]\} \\ &-8P_\eta(\sin 2\theta)(\sin^2\theta)((\cos\theta)\{(L'_1+S'_1)[(\sin^2\theta)L''_2+S''_2]-(L''_1+S''_1)[(\sin^2\theta)L'_2+S'_2]\} \\ &+(\sin^3\theta)(S'_3L''_2-S''_3L'_2)) \\ &+8P_\xi(\sin 2\theta)(\sin^2\theta)((\sin^2\theta)(\cos\theta)[(L'_1+S'_1)L'_2+(L''_1+S''_1)L''_2] \\ &+(\sin\theta)\{S'_3[(\sin^2\theta)L'_2+S'_2]+S''_3[(\sin^2\theta)L''_2+S''_2]\})]\ . \end{aligned} \tag{14}$$

In this expression prime and double prime refer to the real and imaginary parts, respectively, of the generally complex structure factors **L** and **S**. Unprimed variables L_j and S_j refer to the jth components of the complex structure factors, $L_j = L'_j + iL''_j$ and $S_j = S'_j + S''_j$, with i and j labeling different symmetry directions in the U basis (see Fig. 1). When $i \neq j$ and $P \neq 0$ terms of the form L_iS_j, L_iL_j, and S_iS_j occur. In many materials the components associated with different symmetry directions are equal, so that $S_iS_j \to |S_i|^2$. The magnetic structure of erbium,[2] however, is an example where for intermediate temperatures the c axis and basal plane magnetic structures are distinct—thereby giving rise to just this sort of interference in the pure magnetic scattering. It may also be seen from this expression that for **L** and **S** purely real or imaginary, the cross section is independent of the circular component. For a structure factor whose spatial direction is parallel to a U-basis vector, the cross section is independent of both the circular and 45°-linear components.

C. Interference scattering

From Eq. (8) we write

$$\begin{aligned}\left.\frac{d\sigma}{d\Omega}\right|_I &= -\left[\frac{i\hbar\omega}{mc^2}\right]\left[\frac{e^2}{mc^2}\right]^2 \mathrm{tr}(\langle M_m\rangle\rho\langle M_c^\dagger\rangle - \langle M_c\rangle\rho\langle M_m^\dagger\rangle) \\ &= \left[\frac{e^2}{mc^2}\right]^2\frac{\hbar\omega}{mc^2}[\mathrm{Im}(\rho^* m_{11}+\rho^* m_{22}\cos 2\theta)+P_\zeta \mathrm{Im}(\rho^* m_{11}-\rho^* m_{22}\cos 2\theta)+P_\xi \mathrm{Im}(\rho^* m_{12}+\rho^* m_{21}\cos 2\theta) \\ &\quad +P_\eta \mathrm{Re}(\rho^* m_{12}-\rho^* m_{21}\cos 2\theta)]\ , \end{aligned} \tag{15}$$

where we have substituted for $\langle M_c \rangle$ using Eq. (7) and $m_{i,j}$ represent the elements of $\langle M_m \rangle$. When $\langle M_m \rangle$ is diagonal the interference term is independent of the circular and the 45°-linear components. When $\langle M_m \rangle$ is off diagonal, the only coupling is to the circular and the 45°-linear components.

Substituting for $\langle M_m \rangle$ from Eq. (6),

$$\left.\frac{d\sigma}{d\Omega}\right|_I = \left[\frac{e^2}{mc^2}\right]^2 \frac{\hbar\omega}{mc^2}((\sin 2\theta)(1+P_\zeta)(\rho' S_2'' - \rho'' S_2')$$
$$+(\sin 2\theta)(\cos 2\theta)(1-P_\zeta)\{\rho'[2(\sin^2\theta)L_2''+S_2'']-\rho''[2(\sin^2\theta)L_2'+S_2']\}$$
$$-P_\eta(\sin\theta)\{(1+\cos 2\theta)(\sin 2\theta)[\rho'(L_1'+S_1')-\rho''(L_1''+S_1'')]-(1-\cos 2\theta)^2(\rho' S_3'-\rho'' S_2'')\}$$
$$-P_\xi(\sin\theta)\{(1-\cos 2\theta)(\sin 2\theta)[\rho'(L_1''+S_1'')-\rho''(L_1'+S_1')]$$
$$-(1+\cos 2\theta)(1-\cos 2\theta)(\rho' S_3''-\rho'' S_3')\}) . \quad (16)$$

In this expression ρ' and ρ'' refer to the real and imaginary parts of the charge form factor, respectively. When $\mathbf{P}=0$ the only contributions to the interference term come from magnetization density oriented perpendicular to the diffraction plane. The interference term is considerably simplified for centrosymmetric systems (when $L_j''=S_j''=0$). Then,

$$\left.\frac{d\sigma}{d\Omega}\right|_I = -\left[\frac{e^2}{mc^2}\right]^2 \frac{\hbar\omega}{mc^2}\{(\sin 2\theta)(1+P_\zeta)\rho'' S_2'' + (\sin 2\theta)(\cos 2\theta)(1-P_\zeta)\rho''[2(\sin^2\theta)L_2+S_2]$$
$$+4(\sin^2\theta)P_\eta[(\cos^3\theta)\rho'(L_1+S_1)-(\sin^3\theta)\rho' S_3]$$
$$-4(\sin^2\theta)P_\xi[(\sin^2\theta)(\cos\theta)\rho''(L_1+S_1)-(\sin\theta)(\cos^2\theta)\rho'' S_2]\} . \quad (17)$$

It is seen in this case that the circular component couples to the real part of the electronic form factor, while both linear components couple to the imaginary part of the electronic form factor.

V. EXAMPLES

We now apply the general formulas obtained above to several simple examples of magnetic structures. We recall in this regard that synchrotron radiation is predominantly linearly polarized within the median plane of the storage ring ($P_\zeta=\pm 1$, depending on the orientation of the diffraction plane) and elliptically polarized above and below the median plane[19] ($P_\eta, P_\zeta \neq 0$). The detailed polarization dependence of the incident beam depends on a number of machine parameters including beam size, magnet geometry, electron energy, etc.

A. Ferromagnets

In ferromagnets the magnetic and charge scattering are coincident in reciprocal space. Since the magnetic scattering is typically reduced from the charge scattering by $> \sim 10^{-6}$ it is difficult to measure the magnetic scattering from ferromagnets directly. One method to over come this limitation is to introduce a magnetic field and measure the flipping ratio, thereby isolating the interference term in the cross section.[4,6] As will be seen, it is also possible to isolate the interference term by "flipping" the incident polarization.[5] In addition, because the magnetic scattering also flips the incident polarization (for some geometries), it is in principle possible to measure the pure magnetic scattering from a ferromagnet by analyzing the polarization of the scattered beam. Experiments performed to analyze the polarization of the magnetically scattered beam are described in references 10 and 20. In the Appendix we develop a formalism for these experimental schemes by introducing the D matrix for detection efficiency. For written simplicity we assume below that the spin- and orbital-angular-momentum densities are collinear.

(i) If $\mathbf{L}$ and $\mathbf{S}$ are perpendicular to the diffraction plane (parallel to $\hat{U}_2$), then

$$\langle M_m \rangle = \sin(2\theta)\begin{vmatrix} S & 0 \\ 0 & 2(\sin^2\theta)L+S \end{vmatrix}$$

and the interference term is

$$\left.\frac{d\sigma}{d\Omega}\right|_I = \left[\frac{e^2}{mc^2}\right]^2\left[-\frac{\hbar\omega}{mc^2}\right](\sin 2\theta)((1+P_\zeta)(\rho' S''-\rho'' S')$$
$$+(1-P_\zeta)(\cos 2\theta)\{\rho'[2(\sin^2\theta)L''+S'']-\rho''[2(\sin^2\theta)L'+S']\}) ,$$

independent of P_ξ and P_η. In this case the real and imaginary parts of the charge form factor multiply the imaginary and real parts of the magnetic structure factor, respectively. The interference cross sections for purely circular and 45°-linear incident polarizations are identical and equal to the result for $\mathbf{P}=0$. Note that for purely linear incident polarization ($P_\zeta=\pm1$) it is possible to separate $\mathbf{L}$ and $\mathbf{S}$ by alternately scattering in the horizontal and vertical planes. For centrosymmetric systems with $P_\zeta=1$ we recover the simple result[4,6]

$$\left.\frac{d\sigma}{d\Omega}\right|_I=\left[\frac{e^2}{mc^2}\right]^2\left[\frac{-2\hbar\omega}{mc^2}\right](\sin 2\theta)\rho''S\ .$$

The magnetic scattering for $\mathbf{L}$ and $\mathbf{S}$ along $\hat{\mathbf{U}}_2$ is

$$\left.\frac{d\sigma}{d\Omega}\right|_m=\left[\frac{e^2}{mc^2}\right]^2\left[\frac{\hbar\omega}{mc^2}\right]^2\frac{1}{2}(\sin^2 2\theta)[(1+P_\zeta)\,|S|^2+(1-P_\zeta)\,|2(\sin^2\theta)L+S|^2]\ ,$$

independent of P_ξ and P_η. The final polarizations are

$$P'_\xi=(P_\xi\{S'[2(\sin^2\theta)L'+S']+S''[2(\sin^2\theta)L''+S'']\}$$
$$-P_\eta\{S'[2(\sin^2\theta)L''+S'']-S''[2(\sin^2\theta)L'+S']\})/\tfrac{1}{2}[(1+P_\zeta)\,|S|^2+(1-P_\zeta)\,|2(\sin^2\theta)L+S|^2]\ ,$$

$$P'_\eta=(-P_\xi\{S''[2(\sin^2\theta)L'+S']-S'[2(\sin^2\theta)L''+S'']\}$$
$$+P_\eta\{S'[2(\sin^2\theta)L'+S']+S''[2(\sin^2\theta)L''+S'']\})/\tfrac{1}{2}[(1+P_\zeta)\,|S|^2+(1-P_\zeta)\,|2(\sin^2\theta)L+S|^2]\ ,$$

$$P'_\zeta=\frac{(1+P_\zeta)\,|S|^2-(1-P_\zeta)\,|2(\sin^2\theta)L+S|^2}{(1+P_\zeta)\,|S|^2+(1-P_\zeta)\,|2(\sin^2\theta)L+S|^2}\ .$$

For noncentrosymmetric systems the magnetic scattering mixes the 45°-linear and circular components. Note also that for $\mathbf{L}=0$,

$$\frac{d\sigma}{d\Omega}=\left[\frac{e^2}{mc^2}\right]^2\left[\frac{\hbar\omega}{mc^2}\right]^2(\sin^2 2\theta)\,|S|^2,\quad \mathbf{P}'=\mathbf{P}\ .$$

Thus, in this configuration it is the orbital magnetization density which introduces the polarization dependence into the magnetic cross section. Finally, we point out that for unpolarized incident radiation ($\mathbf{P}=0$) and $\mathbf{L}=0$, the magnetic scattering is also unpolarized, $\mathbf{P}'=0$.

(ii) If $\mathbf{L}$ and $\mathbf{S}$ are in the diffraction plane and parallel to $\hat{\mathbf{U}}_1$, then

$$\langle M_m\rangle=-i(\sin 2\theta)(\sin\theta)(L+S)\sigma_\eta\ ,$$

where σ_η is the Pauli matrix defined above. Setting $P_\xi=0$, the interference term is

$$\left.\frac{d\sigma}{d\Omega}\right|_I=\left[\frac{e^2}{mc^2}\right]\left[\frac{-\hbar\omega}{mc^2}\right](\sin^2 2\theta)(\cos\theta)P_\eta$$
$$\times[\rho'(L'+S')-\rho''(L''+S'')]\ .$$

In contrast to the last example, the interference scattering now depends on the product of the real parts of the charge and magnetic structure factors, on the product of the imaginary parts of the charge and magnetic structure factors, on the degree of circular polarization, and on the sum $(\mathbf{L}+\mathbf{S})$. Because of the linear dependence on P_η it is possible to isolate the interference term by taking the difference in intensities above and below the median plane of the storage ring (as well as by measuring flipping ratios in a magnetic field) as has been demonstrated in a powdered ferrite.[5]

The magnetic scattering is given by

$$\left.\frac{d\sigma}{d\Omega}\right|_m=\left[\frac{e^2}{mc^2}\right]\left[\frac{\hbar\omega}{mc^2}\right]^2\sin^2 2\theta\,\sin^2\theta\,|\mathbf{L}+\mathbf{S}|^2\ ,$$

independent of incident polarization. The final polarization is

$$\mathbf{P}'=(P_\xi,-P_\eta,-P_\zeta)\ .$$

The magnetic scattering flips the linear and 45°-linear components, but not the circular component.[21] This suggests that for purely linear incident polarization $P_\zeta=1$ the magnetic scattering may be directly measured by analyzing the rotated linear component.

(iii) If $\mathbf{L}$ and $\mathbf{S}$ are in the diffraction plane[10,22] parallel to $-\hat{\mathbf{U}}_3$, then

$$\langle M_m\rangle=2(\sin^3\theta)S\sigma_\xi\ ,$$

where σ_ξ is the Pauli matrix defined above. The magnetic scattering is

$$\left.\frac{d\sigma}{d\Omega}\right|_m=\left[\frac{e^2}{mc^2}\right]^2\left[\frac{\hbar\omega}{mc^2}\right]^2 4\,|S|^2\sin^6\theta\ ,$$

independent of the incident polarization and orbital-angular-momentum density. In contrast to neutron scattering, the cross section for magnetic x-ray scattering is nonzero when the momentum transfer and the magnetization are collinear. The final polarization is

$$\mathbf{P}'=(P_\xi,-P_\eta,-P_\zeta)\ .$$

The magnetic scattering flips the initial circular and linear polarizations, again suggesting the possibility of measuring it directly by analyzing the polarization of the scattered beam. Setting $P_\xi=0$, the interference term is

$$\left.\frac{d\sigma}{d\Omega}\right|_I=\left[\frac{e^2}{mc^2}\right]^2\left[\frac{\hbar\omega}{mc^2}\right]4(\sin^5\theta)P_\eta(\rho'S'-\rho''S'') .$$

The interference term is again linear in P_η and independent of the orbital-angular-momentum density.

We remark that, in principle, the spin and orbital magnetic-moment contributions to the cross section may each be separately measured in ferromagnets. For example, in cases (ii) and (iii) above (and assuming a centrosymmetric system), by rotating the moments from $\hat{\mathbf{U}}_1$ to $\hat{\mathbf{U}}_3$, and measuring flipping ratios in each direction, the interference scattering is first proportional to $L+S$ and then to S. The directions $\hat{\mathbf{U}}_1$ and $\hat{\mathbf{U}}_3$ are particularly convenient as the ratio R of the two cross sections is also simple,

$$R=\frac{(\sin\theta)^3}{\cos\theta}\frac{S}{L+S} .$$

Similarly, by analyzing the final polarization for two directions of the moment, the pure magnetic scattering may also be used to separate L and S. Provided the incident polarization is well characterized, these same techniques apply to the directions $\hat{\mathbf{U}}_1$ and $\hat{\mathbf{U}}_2$.

It is also worth commenting that the angular and polarization dependence of the magnetic and interference scattering and the final polarization of the scattered beam may all be used to determine unknown ferromagnetic structures. For example, the existence of interference or magnetic scattering at chemical Bragg positions for linearly polarized incident radiation requires a component of the moment parallel to $\hat{\mathbf{U}}_2$. Similarly, the existence of magnetic or interference scattering for circularly polarized incident radiation requires that magnetization density lie in the $\hat{\mathbf{U}}_1$-$\hat{\mathbf{U}}_3$ plane. These directions may be distinguished by studying the angular dependence of the cross section for several different reflections or by analyzing the final polarization. A general technique for determining unknown magnetic structures by x-ray scattering is to study the angular dependence of the magnetic or interference cross sections, Eq. (14) and (16), for rotations of the sample about the momentum transfer. Although these last remarks have been made in a discussion of ferromagnetic structures, similar statements are possible for antiferromagnetic structures, and particularly for uniaxially modulated structures.

B. Antiferromagnets

In the following we make an analogy to the rare-earth elements and write the structure factors for $\mathbf{L}(\mathbf{K})$ and $\mathbf{S}(\mathbf{K})$ in terms of a single quantum $\mathbf{J}=\mathbf{L}+\mathbf{S}$. Thus,

$$\mathbf{L}(\mathbf{K})=\phi_l(\mathbf{K})\sum_{n\ \text{atoms}}^{\text{unit cell}}\mathbf{J}_n(0)e^{i\mathbf{K}\cdot\mathbf{n}}$$

and

$$\mathbf{S}(\mathbf{K})=\phi_s(\mathbf{K})\sum_{n\ \text{atoms}}^{\text{unit cell}}\mathbf{J}_n(0)e^{i\mathbf{K}\cdot\mathbf{n}} ,$$

where

$$\phi_l(\mathbf{K})=\frac{\frac{1}{2}\left\langle a\left|\mathbf{J}\cdot\sum_j^{\text{atom}}[\mathbf{L}_j(0)f(\mathbf{K}\cdot\mathbf{r}_j)+f(\mathbf{K}\cdot\mathbf{r}_j)\mathbf{L}_j(0)]\right|a\right\rangle}{(\frac{1}{2}\mathbf{L}\cdot\mathbf{J}+\mathbf{S}\cdot\mathbf{J})} ,$$

$$\phi_s(\mathbf{K})=\frac{\left\langle a\left|\mathbf{J}\cdot\sum_j^{\text{atom}}\mathbf{s}_je^{i\mathbf{K}\cdot\mathbf{r}_j}\right|a\right\rangle}{(\frac{1}{2}\mathbf{L}\cdot\mathbf{J}+\mathbf{S}\cdot\mathbf{J})},\quad \mathbf{J}_n=J\sum_i g_i(\boldsymbol{\tau}\cdot\mathbf{n})\hat{\mathbf{U}}_n(i) .$$

In these expressions $\mathbf{n}$ is the vector giving the position of the nth atom in a magnetic unit cell, $\boldsymbol{\tau}$ is the modulation wavevector, g_i gives the ith component of the magnetization in the $\hat{\mathbf{U}}$ basis, and $\hat{\mathbf{U}}_n(i)$ specifies the direction of the ith component of the magnetization of the nth atom. $\phi_s(\mathbf{K})$ and $\phi_l(\mathbf{K})$ are the ionic form factors for the spin- and orbital-angular-momentum densities, respectively.[15,22]

(i) For uniaxially modulated systems, $\mathbf{J}_n=Jg(\boldsymbol{\tau}\cdot\mathbf{n})\hat{\mathbf{U}}_i$ (g periodic) and all the results derived above for ferromagnets apply by introducing the prefactor

$$\left|\sum g(\boldsymbol{\tau}\cdot\mathbf{n})e^{i(\mathbf{K}-\boldsymbol{\tau})\cdot\mathbf{n}}\right|^2J^2$$

and by making the replacements $S_i\to\phi_s$ and $L_i\to\phi_l$. In contrast to the case for ferromagnets, the magnetic scattering is now located at positions distinct from the charge scattering and so may be directly measured. It follows, of course, that the interference scattering is zero. By measuring the cross section of the magnetic scattering for suitable orientations of the moments (for example, by rotation of the sample about the momentum transfer or by use of a magnetic field), detailed magnetic-structure determinations and separation of the orbital and spin form factors are possible in a manner analogous to that in ferromagnets.

(ii) The final example we consider is that of a simple basal plane spiral with modulation wavevector $\boldsymbol{\tau}$ oriented along $\hat{\mathbf{U}}_3$, $\boldsymbol{\tau}=\tau\hat{\mathbf{U}}_3$. In that case we have

$$\mathbf{J}_n=[\cos(\boldsymbol{\tau}\cdot\mathbf{n})\hat{\mathbf{x}}+\sin(\boldsymbol{\tau}\cdot\mathbf{n})\hat{\mathbf{y}}]$$

$$=\frac{J}{2}(\hat{\mathbf{U}}_+e^{-i\boldsymbol{\tau}\cdot\mathbf{n}}+\hat{\mathbf{U}}_-e^{i\boldsymbol{\tau}\cdot\mathbf{n}}) ,$$

where $\hat{\mathbf{U}}_+=\hat{\mathbf{U}}_1+i\hat{\mathbf{U}}_2$ and $\hat{\mathbf{U}}_-=\hat{\mathbf{U}}_1-i\hat{\mathbf{U}}_2$. Then $M_m\to M_m^++M_m^-$, giving magnetic scattering at satellites split symmetrically about each chemical Bragg peak along the direction of $\hat{\mathbf{U}}_3$. Thus

$$\langle M_m^\pm\rangle=\frac{J}{2}\sum_n^{\text{cell}}e^{i(\mathbf{K}\mp\boldsymbol{\tau})\cdot\mathbf{n}}(\sin2\theta)$$

$$\times\begin{bmatrix}\pm i\phi_s & -\sin\theta(\phi_l+\phi_s)\\ (\sin\theta)(\phi_l+\phi_s) & \pm i[2(\sin^2\theta)\phi_l+\phi_s]\end{bmatrix} .$$

For simplicity we specialize to the case of linear polarization with $P_\zeta=1$ and assume ϕ_s and ϕ_l are real. Then the magnetic scattering may be written

$$\left.\frac{d\sigma}{d\Omega}\right|_m=\left[\frac{e^2}{mc^2}\right]^2\left[\frac{\hbar\omega}{mc^2}\right]^2\left|\sum_n e^{i(\mathbf{K}\mp\tau)\cdot\hat{\mathbf{n}}}\right|^2\left|\frac{J}{2}\right|^2$$
$$\times(\sin^2 2\theta)(\,|\phi_s|^2+|\phi_l+\phi_s|^2\sin^2\theta)$$

and the final polarization is

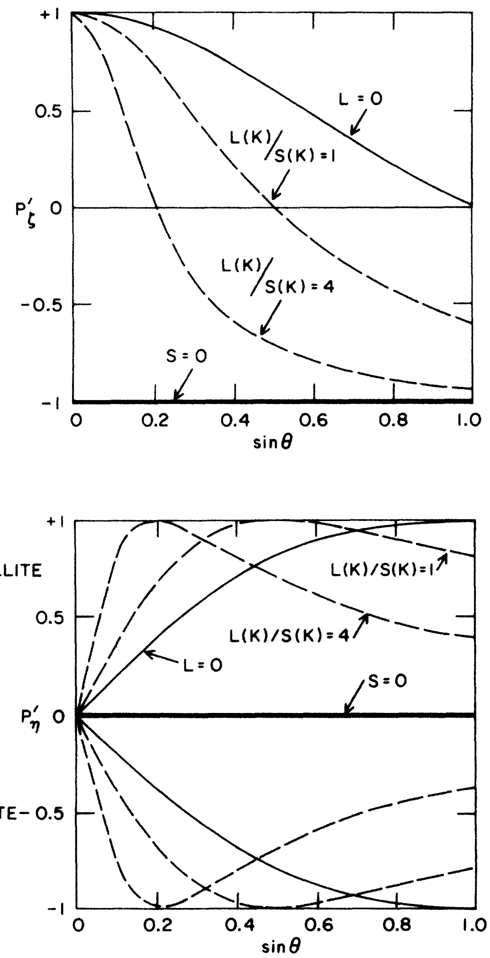

FIG. 3. K dependence of the scattered linear and circular Poincaré coefficients for linearly polarized radiation ($P_\zeta=1$) incident upon a magnetic spiral. Upper: When $\mathbf{L}=0$, then $P'_\zeta=[(1-\sin^2\theta)/(1+\sin^2\theta)]$ is positive definite and decreases from 1 to 0 with increasing momentum transfer $\mathbf{K}$. When $\mathbf{S}=\mathbf{0}$ (and $\theta>0$), then $P'_\zeta=-1$. The dashed lines illustrate the general behavior for two simple cases when the ratio of orbital to spin form factor is constant. Lower: When $\mathbf{L}=\mathbf{0}$ the scattered circular Poincaré coefficient for the positive satellite is negative definite and decreases from 0 to -1 $[P'_\eta=-2\sin\theta/(1+\sin^2\theta)]$. When $\mathbf{S}=0$, then $P'_\eta=0$. The behavior for the negative satellite mirrors that for the positive satellite. For nonzero spin there is always a value of θ for which the scattered beam may be totally circular.

$$P'_\xi=0\ ,$$
$$P'_\eta=\frac{\mp 2(\sin\theta)\phi_s(\phi_l+\phi_s)}{|\phi_s|^2+|\phi_l+\phi_s|^2\sin^2\theta}\ ,$$
$$P'_\zeta=\frac{|\phi_s|^2-|\phi_l+\phi_s|^2\sin^2\theta}{|\phi_s|^2+|\phi_l+\phi_s|^2\sin^2\theta}\ .$$

From this result it is apparent that the term in the cross section proportional to $|\phi_s|^2$ is the probability for polarization parallel to $\hat{\epsilon}_\perp$ in Fig. 1, while the term proportional to $|\phi_l+\phi_s|^2\sin^2\theta$ is that for polarization parallel to $\hat{\epsilon}_\parallel$. Thus, by analyzing the degree of linear polarization in the scattered beam it is possible to separately measure the real form factors $\phi_l(\mathbf{K})$ and $\phi_s(\mathbf{K})$ in a magnetic spiral[20] (see Fig. 3). Provided there is a single spiral domain of well-defined helicity, then the circular components of the positive and negative satellites will have opposite helicity, as shown in Fig. 3. It follows that by adding a circularly polarized component to the incident beam and measuring the degree of linear polarization for the two satellites, the helicity of the spiral may be determined (see Fig. 4). Finally, note that if

$$\sin^2\theta=\frac{|\phi_s(\mathbf{K})|^2}{|\phi_l(\mathbf{K})+\phi_s(\mathbf{K})|^2}\ ,$$

then the scattering is totally circular. Although the efficiency is very low ($<10^{-6}$), it is therefore possible to completely convert linearly polarized radiation to circu-

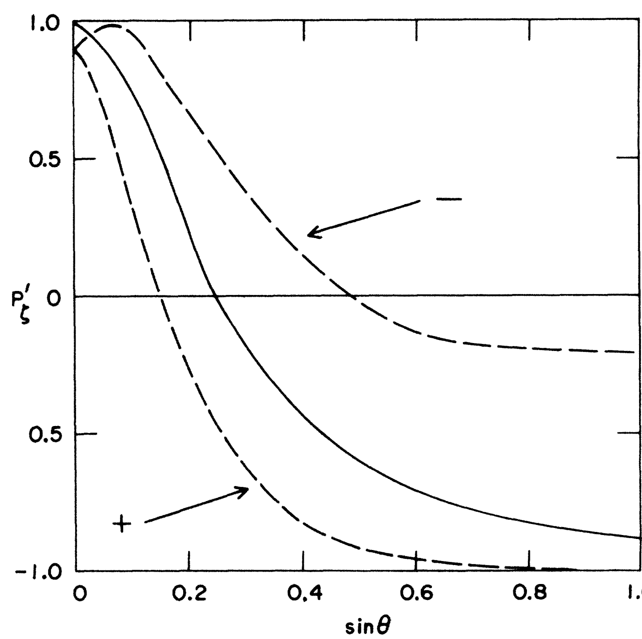

FIG. 4. Linear Poincaré coefficient for scattering from a magnetic spiral for the case $L(K)/S(K)=3$. The solid line shows P'_ζ for incident Poincaré vector $P_\zeta=1$ and $P_\eta=0$. There is no difference between positive and negative satellites. The dashed lines illustrate the change when a small component of circular polarization is introduced in the incident beam ($P_\zeta=0.90$ and $P_\eta=0.43$).

lar by scattering from a magnetic spiral. From theoretical calculations of the form factors for ϕ_l and ϕ_s,[22] it turns out that this condition is approximately satisfied for the $(0\,0\,2\frac{1}{6})$ satellite of holmium with ~ 10 keV incident photon energy.

VI. SUMMARY

In this paper we have derived general expressions for the polarization dependence of magnetic x-ray scattering for high photon energies and indicated a variety of directions for new kinds of synchrotron experiments. The extension of this work into the resonant regime, when the photon energy is near an excitation energy of the solid, will likely produce novel effects and remains to be carried out. Finally, it is worth mentioning that while polarization-dependent magnetic x-ray scattering experiments are possible with present-day synchrotron sources, reliable intensity measurements of the sort required for some of these experiments are still difficult. This class of experiment will clearly benefit from the next generation of synchrotron sources, and particularly by the development of beamlines or insertion devices with tunable polarization characteristics.

Note added in proof. After completion of this manuscript, we received a copy of this work by S. Lovesey [J. Phys. C (in press)].

ACKNOWLEDGMENT

We have benefited from discussions with Alan Goldman and Denis McWhan. One of us (M.B.) would like to acknowledge the hospitality of the Stanford Synchrotron Radiation Laboratory, where some of this work was carried out. Work done at Brookhaven was supported by the Division of Materials Sciences, U.S. Department of Energy under Contract No. DE-AC02-76CH00016.

APPENDIX: D MATRIX FOR DETECTION EFFICIENCY

Quantitative polarization analysis may be included in the formalism by defining a matrix D which represents the detection efficiency and polarization sensitivity of the detector assembly:

$$\frac{d\sigma}{d\Omega}=\left[\frac{e^2}{mc^2}\right]^2 \mathrm{tr}(DM\rho M^\dagger)\ .$$

In this formalism an open detector has the D matrix

$$D_1=e_1 1\ ,$$

where e_1 is the quantum efficiency for the detector. A simple analyzing crystal diffracting within the scattering plans has the D matrix

$$D_2=e_2\begin{bmatrix}1 & 0\\ 0 & \cos^2 2\theta\end{bmatrix}$$

$$=\frac{e_2}{2}(1+\cos^2 2\theta)1+\frac{e_2}{2}(1-\cos^2 2\theta)\sigma_\zeta\ ,$$

where e_2 is the analyzer reflectivity and 2θ its Bragg angle. The D matrix for a linear polarization analyzer oriented at $\phi°$ to the scattering plane[10,20] is

$$D_3=e_3\begin{bmatrix}\cos^2\phi & 0\\ 0 & \sin^2\phi\end{bmatrix}=\frac{e_3}{2}[1+\cos(2\phi)\sigma_\zeta]\ ,$$

where e_3 is the analyzer crystal reflectivity. Operationally, the D matrix for the linear polarization analyzer simply multiplies m_{11} and m_{12} by $\cos\phi$ and multiplies m_{22} and m_{21} by $\sin\phi$.

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