Feasibility of x-ray studies of the magnetic structure of crystals

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The amplitude for the scattering of light by an atom is derived as a function of the atomic angular momentum for frequencies in the intervals $m\alpha^2 \ll \omega \ll m\alpha$ and $m\alpha < \omega \ll m$. The scattering of x rays by a crystal with a magnetic order is analyzed. For a helicoidal spin structure and circularly polarized x rays, the scattering cross section depends on not only the pitch but also the sign of the helix.

INTRODUCTION

The feasibility of using x radiation to determine the magnetic structure of collinear antiferromagnets has been discussed theoretically[1,2] and has in fact been demonstrated experimentally[3,4] for substances with an incomplete d shell. In the present paper we derive an expression for the amplitude for x-ray scattering by an atom or ion as a function of the atomic angular momentum. We point out some new mechanisms for this scattering, which are no less effective than that proposed in Refs. 1 and 2. We analyze the scattering by a magnetically ordered crystal in the particular case of an antiferromagnet with a helicoidal spin structure of the "simple helix" type. We show that the system of magnetic satellites around the Bragg reflections ("ancestors") embodies information about not only the pitch of the helix but also its sign in the case in which the x radiation is circularly polarized.

Although the spin-dependent scattering amplitude is three or four orders of magnitude smaller than the ordinary isotropic amplitude (more on this below), it has been demonstrated experimentally that x radiation can be used to study magnetic structures. The feasibility of such experiments becomes particularly clear when we consider the use of synchrotron radiation, which is incomparably more intense than ordinary x radiation. Studies of magnetic structures with the help of synchrotron radiation may prove an important supplement to neutron-diffraction methods, which are an extremely rich source of information about magnetic order in solids (see Ref. 5, for example). The high collimation of synchrotron radiation may be an important advantage over neutron diffraction in studies of the magnetic structure of samples of very small dimensions. The use of x radiation may also prove extremely convenient for studying substances which exhibit an anomalously high neutron absorption. One final and important advantage of synchrotron radiation is its natural elliptical polarization, whose sign depends on whether the radiation is extracted above or below the orbital plane of the radiating particles in the magnetic field.

ASYMPTOTIC BEHAVIOR OF THE DYNAMIC POLARIZABILITY OF AN ATOM

The amplitude for elastic scattering of light by an isolated atom at the point $\mathbf{R}$ is given in the dipole approximation by the well-known expression[6]

$$f^{E1,\,E1}=\omega^2 e_i'^{*} e_k \alpha_{ik}(\omega)\, e^{-i\varkappa\mathbf{R}}. \tag{1}$$

Here $\varkappa = \mathbf{k}' - \mathbf{k}$, where $\mathbf{k}$ and $\mathbf{k}'$ are the wave vectors of the incident and scattered photons, and $\mathbf{e}$ and $\mathbf{e}'$ are their unit polarization vectors. We are using a system of units with $\hbar = c = 1$. The tensor $\alpha_{ik}(\omega)$ is the dynamic polarizability of an atom in the state $|0\rangle$:

$$\alpha_{ik}(\omega)=\sum_n \left\{ \frac{\langle 0|d_i|n\rangle\langle n|d_k|0\rangle}{\omega_{n0}-\omega} + \frac{\langle 0|d_k|n\rangle\langle n|d_i|0\rangle}{\omega_{n0}+\omega} \right\}, \tag{2}$$

where the operator $\mathbf{d}$ represents the dipole moment, and $\omega_{no} = E_n - E_0$, where E_0 and E_n are the energies of the ground and excited states. If the state $|0\rangle$ of the atom has a total angular momentum $\mathbf{J}$, the polarizability tensor can be written

$$\alpha_{ik}(\omega)=\delta_{ik}\alpha_s(\omega)+i\varepsilon_{ikl}J_l\alpha_v(\omega)+Q_{ik}\alpha_t(\omega),$$
$$Q_{ik}=J_iJ_k+J_kJ_i-2\delta_{ik}J(J+1)/3. \tag{3}$$

The scalar polarizability $\alpha_s(\omega)$, the vector polarizability $\alpha_v(\omega)$, and the tensor polarizability $\alpha_t(\omega)$ have the following properties since the tensor $\alpha_{ik}(\omega)$ is Hermitian:

$$\alpha_{s,v,t}(\omega)=\alpha^{*}_{s,v,t}(\omega),$$
$$\alpha_s(-\omega)=\alpha_s(\omega),\quad \alpha_v(-\omega)=-\alpha_v(\omega),\quad \alpha_t(-\omega)=\alpha_t(\omega). \tag{4}$$

The vector and tensor terms in (3) clearly embody information on the orientation of the total angular momentum of the atom.

We are interested in the scattering of x rays, whose frequencies satsify $\omega \gg \mathrm{Ry} = m\alpha^2/2$, where m is the mass of an electron, and $\alpha = e^2 = 1/137$. We seek the asymptotic behavior of the polarizability in the limit $\omega \to \infty$. The leading term in the expansion of the tensor $\alpha_{ik}(\omega)$ in powers of $1/\omega$ is well known,

$$\alpha_{ik}(\omega\to\infty)\approx-(Z'\alpha/m\omega^2)\,\delta_{ik}, \tag{5}$$

and it determines the scalar polarizability $\alpha_s(\omega)$. Here Z' is the number of outer electrons, for which the characteristic frequencies are much smaller than ω; this number satisfies $Z' \leqslant Z$, where Z is the atomic number.

At first glance it would appear that a vector polarizability could arise only in the next order in $1/\omega$:

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$$\frac{\alpha}{\omega^3}\sum \langle[[\hat{H},r_i],[\hat{H},r_k]]\rangle, \tag{6}$$

where $\hat{H}$ is the Hamiltonian of the atom, and $\mathbf{r}$ is the radius vector of the electron. The summation here is over all the atomic electrons. Expression (6) becomes nonzero only when we incorporate in $\hat{H}$ the spin-orbit interaction, thereby determining the asymptotic behavior of the vector polarizability of the atom. We will show, however, that this contribution to $\alpha_v(\omega\to\infty)$ is not the dominant one. In order to take the relativistic corrections into account systematically we need to redefine the dipole-moment operator in expression (2).

The operator representing the interaction of an electron with an electromagnetic field contains both terms which are linear in the field and terms which are quadratic. When the relativistic corrections are taken into account, these terms become

$$\hat{H}_1=-\frac{e}{2m}[\mathbf{p}\times\mathbf{A}]_+-\frac{e}{2m}\boldsymbol{\sigma}\mathbf{H}+\frac{e}{8m^2}\boldsymbol{\sigma}([\mathbf{p}\times\mathbf{E}]-[\mathbf{E}\times\mathbf{p}]), \tag{7}$$

$$\hat{H}_2=\frac{\alpha}{2m}\mathbf{A}^2-\frac{\alpha}{4m^2}\boldsymbol{\sigma}[\mathbf{A}\times\mathbf{E}]. \tag{8}$$

Here $[.,.]_+$ specifies the anticommutator; $\mathbf{A}$, $\mathbf{E}$, and $\mathbf{H}$ are respectively the vector potential and the electric and magnetic fields;

$$\mathbf{E}=-\partial\mathbf{A}/\partial t,\quad \mathbf{H}=\mathrm{rot}\,\mathbf{A},\quad \mathrm{div}\,\mathbf{A}=0,\quad \mathbf{A}=\mathbf{e}e^{i\mathbf{kr}-i\omega t}; \tag{9}$$

the operator $\mathbf{p}$ represents the electron momentum; and the $\boldsymbol{\sigma}$ are the Pauli matrices. When the relativistic corrections are taken into account, we can use the transformation

$$-\frac{e}{2m}[\mathbf{p},\mathbf{A}]_++\frac{e}{8m^2}\boldsymbol{\sigma}([\mathbf{p}\times\mathbf{E}]-[\mathbf{E}\times\mathbf{p}])$$

$$=-ie\left[\hat{H},\mathbf{rA}+\frac{1}{4m}[\mathbf{r}\times\boldsymbol{\sigma}]\mathbf{E}\right], \tag{10}$$

which holds for $\omega\ll m\alpha$, to reduce the expression for the electric dipole amplitude for scattering to the form in (1), (2), where the dipole-moment operator should now be understood as

$$\mathbf{d}=e[\mathbf{r}+i(\omega/4m)[\mathbf{r}\times\boldsymbol{\sigma}]]. \tag{11}$$

Substituting (11) into (2), we find, in the leading approximation in $1/\omega$,

$$\alpha_{ik}(\omega\to\infty)\approx i\frac{\alpha}{m^2\omega}\varepsilon_{ikl}S_l,\qquad f^{E1,E1}\approx i\frac{\alpha\omega}{m^2}[\mathbf{e}'^*\times\mathbf{e}]\mathbf{S}, \tag{12}$$

where $\mathbf{S}$ is the total spin of the atom. From (12) and (3) we find an expression for the vector polarizability:

$$\alpha_v(\omega\to\infty)=(\alpha/m^2\omega)(g-1), \tag{13}$$

where $g=1+(\mathbf{J}\cdot\mathbf{S})/J(J+1)$ is the Landé factor.

It is not difficult to see that, just as the asymptotic behavior in (5) for the scalar polarizability can be found directly from the first term in expression (8), without transformation (10), the asymptotic behavior in (13) corresponds to the second term in (8).

We now seek the asymptotic behavior of the tensor polarizability of the atom, pursuing the expansion of tensor (2) in powers of $1/\omega$:

$$\alpha_{ik}(\omega\to\infty)\approx-\frac{Z'\alpha}{m\omega^2}\delta_{ik}+\frac{\alpha}{\omega^4}\sum\langle[[\hat{H},r_i],[\hat{H},[\hat{H},r_k]]]\rangle, \tag{14}$$

where $\hat{H}=\mathbf{p}^2/2m+U$ is the one-electron Hamiltonian. Trivial manipulations lead to the following expression for the second term in (14):

$$-\frac{\alpha}{m^2\omega^4}\sum\langle\nabla_i\nabla_k U\rangle. \tag{15}$$

The scalar part of this expression, which reduces to a small correction and to the first term in (14), is of no interest here. For electrons with an orbital angular momentum $l\neq 0$ we can ignore $\langle\Delta U\rangle$ in comparison with $\langle U'/r\rangle$, and for the irreducible part of (15) we find

$$\frac{3\alpha}{m^2\omega^4}\sum\left\langle\left(r_ir_k-\frac{1}{3}r^2\delta_{ik}\right)\frac{U'}{r^3}\right\rangle. \tag{16}$$

Hence the tensor polarizability of a heavy atom with one outer electron ($l\neq 0$) is

$$\alpha_t(\omega\to\infty)=-\frac{3}{4}\frac{\alpha}{m^2\omega^4}\left\langle\frac{1}{r}\frac{dU}{dr}\right\rangle\frac{1}{j(j+1)}, \tag{17}$$

where $\mathbf{j}$ is the total angular momentum of the electron. The tensor polarizability of a heavy atom with several outer electrons is found by evaluating the sum over all equivalent electrons of the incomplete shell (or over all equivalent holes) in (16). We are interested primarily in the ions Tb^{3+}, Dy^{3+}, Ho^{3+}, and Er^{3+}, for which the f shell is more than half-filled. In the ground state we have $J=L+S$ in this case, and the total spin S is, as usual, maximized by virtue of Hund's rule. The asymptotic expression for the tensor polarizability reduces to the following form for these ions:

$$\alpha_t(\omega\to\infty)=Z\alpha^5\frac{m\eta}{\omega^4}\frac{L(7-4S)}{15J(2J-1)}, \tag{18}$$

where the coefficient η is defined by the relation

$$\left\langle\frac{1}{r}\frac{dU}{dr}\right\rangle=Z\alpha^4m^3\eta. \tag{19}$$

Analysis of the fine structure of rare earth ions and various numerical calculations (see Ref. 7, for example) reveal $\eta\sim 10$. In the frequency interval $m\alpha^2\ll\omega\ll m\alpha$, which is studied in Sections 2 and 3, the condition $\alpha_t\ll\alpha_v$ holds.

The asymptotic behavior ω^{-4} of the tensor polarizability was pointed out previously by Manakov and Ovsyannikov.[8]

CONTRIBUTION OF HIGHER-ORDER MULTIPOLES TO THE ASYMPTOTIC BEHAVIOR OF THE SCATTERING AMPLITUDE

Since we are considering the relativistic correction to the electric dipole scattering amplitude, it is quite natural to examine the amplitudes of higher multipolarities, which arise in the same order in the relativistic small parameter. In particular, there is the magnetic scattering discussed in Refs. 2 and 3. It is not difficult to see, however, that the amplitude for electric quadrupole scattering is on the same order of magnitude, as are the interference amplitudes (E 1, M 2), (E 1, E 3), and (M 1, E 2). A multipole expansion up to E 3 and M 2, inclusively, corresponds to a transformation of Hamiltonian (7) to the form

$$\hat{H}_1 = -ie\left[\hat{H},\ (\mathbf{rA}) + \frac{i}{2}(\mathbf{rk})\,(\mathbf{rA}) - \frac{1}{6}(\mathbf{rk})^2(\mathbf{rA})\right]$$
$$-\frac{e}{2m}\mathbf{H}(\mathbf{l}+\boldsymbol{\sigma})$$
$$-i\frac{e}{2m}(\mathbf{rk})\,(\mathbf{H}\boldsymbol{\sigma}) - i\frac{e}{6m}[\,(\mathbf{rk}),\ (\mathbf{Hl})\,]_+. \quad (20)$$

The last two terms in (20) contain, in addition to the amplitude M 2, yet another relativistic contribution to the electric dipole interaction:

$$-\frac{ie\omega}{4m}\left\{[\boldsymbol{\sigma}\times\mathbf{r}] + \frac{1}{3}[\mathbf{l}\times\mathbf{r}] - \frac{1}{3}[\mathbf{r}\times\mathbf{l}]\right\}\mathbf{E}.$$

We of course do not need the relativistic corrections to the amplitudes of high multipolarities. Substituting (20) into the familiar expression for the light scattering amplitude,

$$f = -\sum\left\{\langle\hat{H}_2\rangle - \sum_n\left[\frac{\langle 0|\hat{H}_1'|n\rangle\langle n|\hat{H}_1|0\rangle}{\omega_{n0}-\omega} + \frac{\langle 0|\hat{H}_1|n\rangle\langle n|\hat{H}_1'|0\rangle}{\omega_{n0}+\omega}\right]\right\}, \quad (21)$$

we can derive amplitudes corresponding to the contributions of the various multipolarities. The radiation operator $\hat{H}_1'$ is related to the photon absorption operator $\hat{H}_1$ by

$$\hat{H}_1'(\mathbf{e}'^*,\ \omega,\ \mathbf{k}') = \hat{H}_1(\mathbf{e},\ -\omega,\ -\mathbf{k}).$$

We examined the electric dipole amplitude (E 1, E 1) in the preceding section. The magnetic dipole amplitude (M 1, M 1) is

$$f^{M1,\,M1} = \omega^2[\mathbf{n}'\times\mathbf{e}'^*]_i[\mathbf{n}\times\mathbf{e}]_k\chi_{ik}(\omega), \quad (22)$$

where $\mathbf{n} = \mathbf{k}/\omega$ and $\mathbf{n}' = \mathbf{k}'/\omega$. The dynamic magnetic susceptibility tensor of the atom is

$$\chi_{ik}(\omega) = \frac{\alpha}{4m^2}{\sum}'\left\{\frac{\langle 0|l_i+\sigma_i|n\rangle\langle n|l_k+\sigma_k|0\rangle}{\omega_{n0}-\omega} + \frac{\langle 0|l_k+\sigma_k|n\rangle\langle n|l_i+\sigma_i|0\rangle}{\omega_{n0}+\omega}\right\}. \quad (23)$$

The prime on the summation sign here means, as usual, that the ground state is eliminated from the number of intermediate states [in expression (2) for $\alpha_{ik}(\omega)$, we do not have to be especially concerned about this point because of the selection rules for E 1 transitions]. In the expansion of the tensor $\chi_{ik}(\omega)$ in powers of $1/\omega$, even the first term is nonzero. It can easily be put in the form

$$\chi_{ik}(\omega\to\infty) = -\frac{\alpha}{4m^2\omega}\langle[L_i+2S_i,\ L_k+2S_k]\rangle$$
$$+\frac{\alpha}{4m^2\omega}\sum\{\langle 0|L_i+2S_i|0'\rangle\langle 0'|L_k+2S_k|0\rangle$$
$$-\langle 0|L_k+2S_k|0'\rangle\langle 0'|L_i+2S_i|0\rangle\}, \quad (24)$$

where the state $|0'\rangle$ can differ from $|0\rangle$ only in the projection M of the total angular momentum. The sum is over all M in the state $|0'\rangle$. From (24) we find

$$\chi_{ik}(\omega\to\infty) \approx i\varepsilon_{ikl}J_l\alpha(g-1)(g-2)/4m^2\omega, \quad (25)$$

where we would have $(g-1)(g-2) = LS/J^2$ for the rare earth ions of interest here. Expression (25) describes the asymptotic behavior of the vector magnetic susceptibility. The corresponding contribution to the scattering amplitude is proportional to ω:

$$f^{M1,M1} \approx i\frac{\alpha\omega}{4m^2}[[\mathbf{n}'\times\mathbf{e}'^*][\mathbf{n}\times\mathbf{e}]]\mathbf{J}(g-1)(g-2). \quad (26)$$

Since the magnetic moment operator couples only states with the same L and S but different J, the condition for the applicability of asymptotic expression (26) is unusually liberal: The frequency ω must be much higher than only the fine-structure interval. On the other hand, in accordance with the condition for the multipole expansion for the atoms, we assume $\omega \ll m\alpha$ in this section. In particular, this condition allows us to ignore the well-known diamagnetic contribution of $\hat{H}_2$ to the scattering amplitude. We will also make systematic use of the condition $\omega \ll m\alpha$ further on in the present section, restricting all the amplitudes to terms linear in the frequency.

The asymptotic expression for the quadrupole amplitude (E 2, E 2) can also be found without difficulty:

$$f^{E2,E2} \approx -\frac{\alpha}{4}\omega e_i'^* e_k n_j' n_s\sum\langle[[\hat{H},r_ir_j],[\hat{H},r_kr_s]]\rangle. \quad (27)$$

The summation in this expressin is over all the electrons. After elementary manipulations we find from (27)

$$f^{E2,E2} \approx -i\frac{\alpha\omega}{4m^2}(g-2)\mathbf{J}\{(\mathbf{ne}'^*)[\mathbf{n}\times\mathbf{e}] - (\mathbf{n}'\mathbf{e})[\mathbf{n}\times\mathbf{e}'^*]$$
$$+(\mathbf{nn}')[\mathbf{e}'^*\times\mathbf{e}] + (\mathbf{e}'^*\mathbf{e})[\mathbf{n}'\times\mathbf{n}]\}. \quad (28)$$

The interference of the last two terms in (20) with the nonrelativistic E 1 amplitude gives us

$$f \approx i\frac{\alpha\omega}{2m^2}g\mathbf{J}\{(\mathbf{n}'\mathbf{e})[\mathbf{n}'\times\mathbf{e}'^*] - (\mathbf{ne}'^*)[\mathbf{n}\times\mathbf{e}]\}. \quad (29)$$

As for the interference amplitudes (E 2, M 1) and (E 1, E 3), we note that it is not difficult to see that they are zero in our approximation.

The total scattering amplitude for a photon with a frequency in the interval $m\alpha^2 \ll \omega \ll m\alpha$, which depends on the angular momentum of the atom or ion [the sum of expressions (12), (26), (28), and (29)], is

$$f = i(\alpha\omega/4m^2)(\mathbf{bJ}), \quad (30)$$

where

$$\mathbf{b}=4(g-1)[\mathbf{e}'^*\mathbf{e}]+(g-1)(g-2)[[\mathbf{e}'^*\times\mathbf{n}'][\mathbf{e}\times\mathbf{n}]]$$
$$-(g-2)\{(\mathbf{n}\mathbf{e}'^*)[\mathbf{n}'\times\mathbf{e}]-(\mathbf{n}'\mathbf{e})[\mathbf{n}\times\mathbf{e}'^*]$$
$$+(\mathbf{n}\mathbf{n}')[\mathbf{e}'^*\times\mathbf{e}]+(\mathbf{e}'\mathbf{e})[\mathbf{n}'\times\mathbf{n}]\}$$
$$+2g\{(\mathbf{n}'\mathbf{e})[\mathbf{n}'\times\mathbf{e}'^*]-(\mathbf{n}\mathbf{e}'^*)[\mathbf{n}\times\mathbf{e}]\}. \quad (31)$$

The second term in (31), which corresponds to the magnetic amplitude $f^{M1,M1}$, is usually numerically small in comparison with the other terms. The angular-momenum-dependent amplitude (30) is a fraction $\sim\omega/Z'm$ of the usual scalar amplitude, which has a value

$$f_1\approx-(Z'\alpha/m)(\mathbf{e}'^*\mathbf{e}), \quad (32)$$

according to (1) and (5).

AMPLITUDE FOR SCATTERING BY AN ATOM IN THE CASE $\omega > m\alpha$

The case $m\alpha^2 \ll \omega \ll m\alpha$ was discussed above, but frequencies in the interval $m\alpha < \omega \ll m$ are also of considerable interest. For this interval we cannot use the multipole expansion in the form in which it has been used previously, but the relativistic corrections are still small, so that we can use interaction operators (7) and (8).

We substitute these expressions for $\hat{H}_1$ and $\hat{H}_2$ into (21). The total scattering amplitude can be written as the sum of five components:

$$f_1=-\frac{\alpha}{2m}\sum\langle \mathbf{A}^2\rangle, \quad (33)$$

$$f_2=\frac{\alpha}{4m^2}\sum\langle\sigma[\mathbf{A}\times\mathbf{E}]\rangle, \quad (34)$$

$$f_3=\frac{\alpha}{4m^2}\sum\left[\frac{\langle 0|\sigma\mathbf{H}'|n\rangle\langle n|\sigma\mathbf{H}|0\rangle}{\omega_{n0}-\omega}+\frac{\langle 0|\sigma\mathbf{H}|n\rangle\langle n|\sigma\mathbf{H}'|0\rangle}{\omega_{n0}+\omega}\right], \quad (35)$$

$$f_4=\frac{\alpha}{2m^2}\sum\left[\frac{\langle 0|\mathbf{p}\mathbf{A}'|n\rangle\langle n|\sigma\mathbf{H}|0\rangle}{\omega_{n0}-\omega}+\frac{\langle 0|\sigma\mathbf{H}|n\rangle\langle n|\mathbf{p}\mathbf{A}'|0\rangle}{\omega_{n0}+\omega}+\frac{\langle 0|\sigma\mathbf{H}'|n\rangle\langle n|\mathbf{p}\mathbf{A}|0\rangle}{\omega_{n0}-\omega}+\frac{\langle 0|\mathbf{p}\mathbf{A}|n\rangle\langle n|\sigma\mathbf{H}'|0\rangle}{\omega_{n0}+\omega}\right], \quad (36)$$

$$f_5=\frac{\alpha}{m^2}\sum\left[\frac{\langle 0|\mathbf{p}\mathbf{A}'|n\rangle\langle n|\mathbf{p}\mathbf{A}|0\rangle}{\omega_{n0}-\omega}+\frac{\langle 0|\mathbf{p}\mathbf{A}|n\rangle\langle n|\mathbf{p}\mathbf{A}'|0\rangle}{\omega_{n0}+\omega}\right]. \quad (37)$$

Expressions (33)–(37) take the following form for the interval $m\alpha < \omega \ll m$:

$$f_1=-\frac{\alpha}{m}(\mathbf{e}'^*\mathbf{e})\sum 4\pi(-i)^t Y_{t\tau}^*(\hat{\varkappa})\langle j_t(\varkappa r)\rangle\langle Y_{t\tau}(\hat{\mathbf{r}})\rangle, \quad (38)$$

$$f=f_2+f_3+f_4$$
$$=-i\frac{\alpha\omega}{m^2}\sum 4\pi(-i)^t Y_{t\tau}^*(\varkappa)\langle j_t(\varkappa r)\rangle\left\langle\left(\mathbf{a}\frac{\sigma}{2}\right)Y_{t\tau}(\hat{\mathbf{r}})\right\rangle, \quad (39)$$

$$f_5=-i\frac{\alpha^2}{m\omega}[[\mathbf{e}'^*\times\mathbf{e}]\varkappa]\sum 4\pi(-i)^t Y_{t\tau}^*(\varkappa)[R_t(\varkappa)\langle Y_{t\tau}(\mathbf{r})\mathbf{r}\rangle$$
$$-\frac{i}{2}S_t(\varkappa)\langle Y_{t\tau}(\hat{\mathbf{r}})[\hat{\mathbf{r}}\times\mathbf{l}]\rangle], \quad (40)$$

where $\varkappa=\mathbf{k}'-\mathbf{k}$, $\hat{\varkappa}=\varkappa/\varkappa$, $\hat{\mathbf{r}}=\mathbf{r}/r$; the radial matrix elements

$$R_t(\varkappa)=(m\alpha)^{-1}\langle j_t(\varkappa r)(\partial/\partial r-2/r)\rangle,$$
$$S_t(\varkappa)=(m\alpha)^{-1}\langle j_t(\varkappa r)/r\rangle$$

are dimensionless; $j_t(\varkappa r)$ are the spherical Bessel functions; $Y_{t\tau}$ are the spherical harmonics;

$$\mathbf{a}=[[\mathbf{n}'\times\mathbf{e}'^*][\mathbf{n}\times\mathbf{e}]]-[\mathbf{e}'^*\times\mathbf{e}]+(\mathbf{e}'^*\mathbf{n})[\mathbf{n}\times\mathbf{e}]-(\mathbf{e}\mathbf{n}')[\mathbf{n}'\times\mathbf{e}'^*]; \quad (41)$$

and the summation is over t, τ, and all the atomic electrons. In the interval $m\alpha \ll \omega \ll m$ we can obviously use the estimates

$$f_5\sim\frac{\alpha^2}{m}\ll f\sim\frac{\alpha\omega}{m^2}\ll f_1\sim\frac{\alpha}{m}, \quad (42)$$

so we will ignore the amplitude f_5 below. We might note that both of the amplitudes f and f_1 depend on the orientation of the total angular momentum of the atom (in addition to the term in f_1 for $t=0$). The dependence on the polarization in f_1, however, is trivial, $f_1 \propto (\mathbf{e}'^*\mathbf{e})$ so that the amplitude f, even though small in magnitude, is of major interest for a study of magnetic structure.

For the amplitude f_1 in (38) we must calculate the sum of one-electron matrix elements over the outer shell for $t\neq 0$:

$$\sum\langle lm|Y_{t0}(\hat{\mathbf{r}})|lm\rangle, \quad (43)$$

where m is the z projection of the orbital angular momentum of the electron. In this expression, t is obviously of even parity and bounded ($t\leqslant 2l$). It is convenient to transform from sum (43) to an operator averaged over a state with a given total angular momentum (but not a given orientation) of the atom. In terms of transformation properties, this operator must be an irreducible tensor of rank t (see Ref. 9, for example) constructed from the total-angular-momentum operators. We choose it in the form

$$T_{t\tau}(\mathbf{J})=\{\ldots\{\{\mathbf{J}_1\otimes\mathbf{J}_1\}_2\otimes\mathbf{J}_1\}_3\ldots\otimes\mathbf{J}_1\}_{t\tau}. \quad (44)$$

Here $\{\mathbf{A}_a\otimes\mathbf{B}_a\}_b$ is the irreducible product of rank b of two operators of rank a. The sum (43) and (38) must be replaced by the expression

$$(4\pi)^{-1/2}A^t_{JLl}T_{t\tau}(\mathbf{J}), \quad (45)$$

where, in the case of LS coupling, the coefficient

$$A_{JLl}=(4\pi)^{1/2}\frac{\langle J\|\mathbf{T}_t(\mathbf{L})\|J\rangle\sum\langle lm|Y_{t0}(\mathbf{r})|lm\rangle}{\langle J\|\mathbf{T}_t(\mathbf{J})\|J\rangle\langle LM_L|T_{t0}(\mathbf{L})|LM_L\rangle} \quad (46)$$

$$M_L=\sum m$$

can be calculated. It is not difficult to see that the A^t_{JLl} are independent of the orientation of $\mathbf{J}$; all the information on this orientation is embodied in $T_{t\tau}(\mathbf{J})$. If we treat $\mathbf{J}$ a a classical vector directed along the unit vector $\hat{\mathbf{J}}$, we can write[9]

$$T_{t\tau}(\mathbf{J})=J^t\left[\frac{4\pi t!}{(2t+1)!!}\right]^{1/2}Y_{t\tau}(\hat{\mathbf{J}}).\qquad(47)$$

In a completely analogous way we can transform the amplitude f. The sum

$$\tfrac{1}{2}\sum\langle(\mathbf{a}\boldsymbol{\sigma})Y_{t\tau}(\hat{\mathbf{r}})\rangle\qquad(48)$$

in (39) can be replaced by the following equivalent operator after it is broken up into irreducible parts, and Hund's rule is applied (the spin of the atom is maximized):

$$(4\pi)^{-1/2}\sum_{r,\nu}(-1)^\nu a_{1-\nu}C^{r\,\tau+\nu}_{t\tau\,1\nu}A^{rt}_{JLl}T_{r\,\tau+\nu}(\mathbf{J}).\qquad(49)$$

Here the coefficient[1)]

$$A^{rt}_{JLl}=\left[\frac{3\pi}{S(S+1)}\right]^{1/2}\times\frac{\langle J\|\{\mathbf{T}_t(\mathbf{L})\otimes\mathbf{S}_1\}_r\|J\rangle\sum\langle lm|Y_{t0}(\hat{\mathbf{r}})|lm\rangle}{\langle J\|\mathbf{T}_r(\mathbf{J})\|J\rangle\langle LM_L|T_{t0}(\mathbf{L})|LM_L\rangle}\qquad(50)$$

does not depend on the orientation of $\mathbf{J}$, and the $C^{c\gamma}_{a\alpha b\beta}$ are the Clebsch-Gordan coefficients.

BRAGG SCATTERING BY A MAGNETIC STRUCTURE

At this point we turn from the amplitude for scattering by an individual atom or ion to the amplitude for scattering by a crystal with a definite magnetic structure, e.g., a "simple helix" helicoidal structure, for which we have

$$\mathbf{J}(\mathbf{R}_{ns})=J[\mathbf{m}_1\cos(\mathbf{g}\mathbf{R}_{ns})+\mathbf{m}_2\sin(\mathbf{g}\mathbf{R}_{ns})],\qquad(51)$$

where $\mathbf{m}_1$ and $\mathbf{m}_2$ are orthogonal unit vectors, $\mathbf{m}=\mathbf{m}_1\times\mathbf{m}_2$, $\mathbf{g}$ is the wave vector of the helix $(\mathbf{g}\cdot\mathbf{m})>0$ for a right-handed helix), the index n specifies the unit cell, and s specifies the ions in it. We can transform from the atomic amplitudes (30), (32), (38), and (39) to the amplitudes for scattering by a crystal by summing over all the ions with a weight $\exp(-i\varkappa\mathbf{R}_{ns})$. In the case $\omega\ll m\alpha$ we find

$$F_1=\sum_{n,s}f_1\exp(-i\varkappa\mathbf{R}_{ns})=-\frac{Z'\alpha}{m}(\mathbf{e}'^*\mathbf{e})N_0\sum_{\mathbf{q}}S(\mathbf{q})\delta_{\varkappa,\mathbf{q}},\qquad(52)$$

$$F_2=\sum_{n,s}f\exp(-i\varkappa\mathbf{R}_{ns})=i\frac{\alpha\omega}{8m^2}N_1\sum_q JS(\mathbf{q})\mathbf{b}[\mathbf{m}_1\delta^{(+1)}-i\mathbf{m}_2\delta^{(-1)}],\qquad(53)$$

where $\mathbf{q}$ is a reciprocal-lattice vector, N_τ is the number of unit cells which contribute to the Bragg reflection with a momentum transfer $\mathbf{q}+\tau\mathbf{g}$, and

$$\delta^{(\pm\tau)}=\delta_{\varkappa,\,\mathbf{q}+\tau\mathbf{g}}\pm\delta_{\varkappa,\,\mathbf{q}-\tau\mathbf{g}},$$

$$\delta_{\varkappa,\mathbf{q}}=\begin{cases}1 & \text{at}\quad \varkappa=\mathbf{q},\\ 0 & \text{at}\quad \varkappa\neq\mathbf{q}.\end{cases}$$

For a crystal with a hexagonal close packed lattice (the lattice of the rare earth metals of interest here), the structure factor is

$$S(\mathbf{q})=\sum_s\exp(-i\mathbf{q}\mathbf{R}_s)=2\cos(\mathbf{q}\boldsymbol{\rho}/2),\qquad(54)$$

where the vector $\boldsymbol{\rho}$ connects two ions in the cell of the hcp structure.

In the case $\omega>m\alpha$ the dependence of the atomic amplitudes on the orientation of $\mathbf{J}$ involves only the tensor $T_{t\tau}(\mathbf{J})$. If we direct the z axis along the vector $\mathbf{m}$, we find, using (47) and (51),

$$T_{t\tau}[\mathbf{J}(\mathbf{R}_{ns})]=J^t\beta_{t\tau}\exp(i\tau\mathbf{g}\mathbf{R}_{ns}),\qquad(55)$$

where

$$\beta_{t\tau}=(-1)^{(t+\tau)/2}\left[\frac{t!(t-\tau-1)!!(t+\tau-1)!!}{(2t-1)!!(t-\tau)!!(t+\tau)!!}\right]^{1/2}\qquad(56)$$

for even $t+\tau$ $(t\neq0)$ and $\beta_{t\tau}=0$ for odd values. The amplitude for scattering by a crystal with a simple helix magnetic structure can be written

$$F_3=-\frac{\alpha}{m}(\mathbf{e}'^*\mathbf{e})\sum_{q,t,\tau}(-i)^tS(\mathbf{q})N_\tau\langle j_t(\varkappa r)\rangle J^tA^{t}_{JLl}\beta_{t\tau}\times(4\pi)^{1/2}Y^*_{t\tau}(\hat{\varkappa})\delta_{\varkappa,\mathbf{q}+\tau\mathbf{g}},\qquad(57)$$

$$F_4=-i\frac{\alpha\omega}{m^2}\sum_{q,t,r,\nu}(-i)^tS(\mathbf{q})N_\nu\langle j_t(\varkappa r)\rangle J^rA^{rt}_{JLl}\beta_{r\nu}\times(4\pi)^{1/2}(\mathbf{a}\mathbf{Y}^{t*}_{r\nu}(\hat{\varkappa}))\delta_{\varkappa,\mathbf{q}+\nu\mathbf{g}},\qquad(58)$$

where $Y^t_{r\nu}(\hat{\varkappa})$ is a vector spherical harmonic.[9]

Since the indices t, τ $(t,|\tau|\leqslant 2l)$ in (57) and (58) take on only even values, while the indices r, ν $(r,|\nu|\leqslant 2l+1)$ take on odd values (see the discussion above), the amplitudes F_3 and F_4 do not interfere in crystals whose magnetic structures are incommensurable with their crystal structures (in which the $\mathbf{q}$'s are not multiples of $\mathbf{g}$).[2)] There is no interference of the amplitudes F_1 and F_2 in this case. In (57) and (58), however, the terms with identical τ or ν, but with differences in other indices, can interfere with each other. The terms with $|\tau|=t=2l$ in (57) and with $|\nu|=r=2l+1$ in (58) do not interfere with any other terms.

In (57) there is an isotropic term

$$F_3(t=0)=-\frac{\alpha}{m}(\mathbf{e}'^*\mathbf{e})N_0\sum_{\mathbf{q}}S(\mathbf{q})\delta_{\varkappa,\mathbf{q}}\sum\langle j_0(\varkappa r)\rangle,\qquad(59)$$

which contains a sum over all the electrons of the ion. In the case $\varkappa r\ll1$, this term is the same as the amplitude F_1. Amplitude (59) at $\omega>m\alpha$ or amplitude (52) at $\omega\ll m\alpha$ determines ordinary Bragg scattering with a momentum transfer $\varkappa=\mathbf{q}$ (Bragg reflections). In the region $\omega\ll m\alpha$, the Bragg reflections acquire a pair of satellites $(\varkappa=\mathbf{q}\pm\mathbf{g})$ with relative intensities

$$(\alpha^2/Z')^2\ll|F_2|^2/|F_1|^2<(\alpha/Z')^2.$$

If, on the other hand, $\omega>m\alpha$, amplitudes (57) and (58) give rise to satellites around the Bragg reflections up to the $2l+1$)st order (we recall that for rare earth ions we have $l=3$). The intensity of the satellites of even order is determined by the square of amplitude (57), while that of the satellites of odd order is determined by the square of amplitude (58). In particular, this result means that the polarization dependence of the intensity of the even satellites is trivial, and there is a relative suppression of the odd satellites:

$$\alpha^2 < |F_4|^2/|F_3|^2 \ll 1.$$

INTENSITY OF THE $(2l+1)$st PAIR OF SATELLITES

As an example we seek the intensity of the $(2l+1)$st pair of the satellites which appear around a Bragg reflection upon scattering from a simple helix magnetic structure.

Taking into account the discussion in the preceding section, we see that the scattering cross section is determined by the square of amplitude (58) with $l=3$:

$$|F_4|^2 \approx \frac{\alpha^2\omega^2}{m^4} N_7^2 \sum_{\mathbf{q}} 4\pi S^2(\mathbf{q}) \langle j_6(\varkappa r)\rangle^2 [A^{76}_{JL3}]^2 J^{14}$$

$$\times\{a^*_{1-1} a_{1-1} \beta_{77}{}^2 Y^*_{66}(\hat{\varkappa}) Y_{66}(\hat{\varkappa}) \delta_{\varkappa,\mathbf{q}+7\mathbf{g}} + a^*_{11} a_{11} \beta^2_{7-7} Y^*_{6-6}(\hat{\varkappa}) Y_{6-6}(\hat{\varkappa}) \delta_{\varkappa,\mathbf{q}-7\mathbf{q}}\}, \quad (60)$$

where we have used $C^{k+1,k+1}_{kk11} = C^{k+1,-k-1}_{k-k1-1} = 1$. Also using (56) and

$$|Y_{kk}(\hat{\varkappa})|^2 = |Y_{k-k}(\hat{\varkappa})|^2 = \frac{(2k+1)!!}{4\pi(2k)!!}\sin^{2k}\theta = \frac{(2k+1)!!}{4\pi(2k)!!}[1-(\hat{\varkappa}\mathbf{m})^2]^k$$

we find

$$|F_4|^2 \approx \frac{\alpha^2\omega^2}{m^4} N_7^2 \frac{15!!}{2^7\cdot 14!!} \sum_{\mathbf{q}} S^2(\mathbf{q})$$

$$\times\langle j_6(\varkappa r)\rangle^2 [A^{76}_{JL3}]^2 J_{14} [1-(\hat{\varkappa}\mathbf{m})^2]^6 \times(a^*_{1-1} a_{1-1} \delta_{\varkappa,\mathbf{q}+7\mathbf{g}} + a^*_{11} a_{11} \delta_{\varkappa,\mathbf{q}-7\mathbf{g}}). \quad (61)$$

Finally, transforming to the Cartesian components of the vector $\mathbf{a}$, and substituting in the values of A^{76}_{JL3}, we find the following result, which applies to the metals of interest here:

$$|F_4|^2 \approx \frac{\alpha^2\omega^2}{m^4} \frac{3S}{(S+1)} \left[\frac{5!7!J^7(2J-7)!N_7}{3!3!2^7(2J)!(2S-1)!(6-2S)!}\right]^2 \times\sum_{\mathbf{q}} S^2(\mathbf{q}) \langle j_6(\varkappa r)\rangle^2$$

$$\times[1-(\hat{\varkappa}\mathbf{m})^2]^6 a_i^* a_j [(\delta_{ij} - m_i m_j)\delta^{(+7)} - i\varepsilon_{ijl} m_l \delta^{(-7)}]. \quad (62)$$

The incident photons are conveneintly described by a polarization density matrix

$$\rho_{ik} = \frac{1}{2}(\delta_{ik} - n_i n_k) + \frac{\xi_1}{2}(e_{1i} e_{2k} + e_{2i} e_{1k}) - i\frac{\xi_2}{2}\varepsilon_{ikl} n_l + \frac{\xi_3}{2}(e_{1i} e_{1k} - e_{2i} e_{2k}), \quad (63)$$

where ξ_1, ξ_2, and ξ_3 are the Stokes parameters, and $\mathbf{e}_1$ and $\mathbf{e}_2$ are orthogonal unit vectors ($[e_1 e_2] = n$). We are not interested in the polarization of the scattered photons. Substituting (63) into (62) in the case $\mathbf{n}\|\mathbf{m}$, we find

$$|F_4|^2 \approx \frac{\alpha^2\omega^2}{m^4} \frac{3S}{(S+1)} \left[\frac{5!7!J^7(2J-7)!N_7}{3!3!2^{10}(2J)!(2S-1)!(6-2S)!}\right]^2 \times\sum_{\mathbf{q}} S^2(\mathbf{q}) \langle j_6(\varkappa r)\rangle^2 (1+x)^6 (1-x)$$

$$\times\{[(1+x)(2-x) + 2\xi_1(\mathbf{e}_1\mathbf{n}')(\mathbf{e}_2\mathbf{n}') + \xi_3[(\mathbf{e}_1\mathbf{n}')^2 - (\mathbf{e}^2\mathbf{n}')^2]]\delta^{(+7)} + \xi_2(1-x^2)\delta^{(-7)}\}, \quad (64)$$

where $x = (\mathbf{n}'\cdot\mathbf{n})$. It follows in particular from (64) that if the degree of circular polarization of the incident photons, ξ_2, is comparable to unity then a change in the sign of the circular polarization or in the sign of the helix will result in a change in the intensity of a given satellite by an amount on the order of unity (a change in the sign of the helix corersponds to a change in the sign of $\mathbf{g}$). Consequently, by making use of the circular polarization of synchrotron radiation one could determine the sign of the magnetic helix in a single-domain sample or the ratio of volumes occupied by domains of different signs.

The intensity of the $(2l+1)$st pair of satellites is conveniently compared with the intensity of the $(2l)$th pair, for which we find the following result for the polarization-independent part from (57) with (56), in the case $\mathbf{n}\|\mathbf{m}$:

$$|F_3|^2 \approx \frac{\alpha^2}{m^2} \left[\frac{5!7!J^6(2J-6)!N_6}{3!3!2^9(2J)!(2S-1)!(6-2S)!}\right]^2 \times\sum_{\mathbf{q}} S^2(\mathbf{q}) \langle j_6(\varkappa r)\rangle^2 \cdot \frac{1}{2}(1+x)^6(1+x^2)\delta^{(+6)}. \quad (65)$$

Here

$$\frac{|F_4|^2}{|F_3|^2} \approx \frac{\omega^2}{m^2} \frac{(1-x^2)(2-x)}{(1+x^2)} \frac{3SJ^2}{2(S+1)(2J-6)^2} \frac{N_7^2}{N_6^2} \sim \frac{\omega^2}{m^2}$$

in accordance with the estimate above.

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Note added in proof (2 July 1984). The dependence of the forward scattering amplitude on the photon helicity λ in the interval $m\alpha^2 \ll \omega \ll m\alpha$ can easily be derived by setting $\mathbf{n}' = \mathbf{n}$ and $\mathbf{e}' = \mathbf{e}$ in (30) and (31):

$$f\lambda = -\frac{g^2}{4}\frac{\alpha\omega}{m^2}(\mathbf{nJ})\lambda.$$

This dependence gives rise to an optical activity of ferromagnets in this frequency range. Although the effect reaches a magnitude of 10^{-1} to 1 rad/cm, it would be extremely difficult to observe because of the strong x-ray absorption.

[1)]It can be shown that we have $A^{\tau}_{JLl} \neq 0$ only at $r = t \pm 1$ in the case $J = L + S$.

[2)]This assertion does not apply to a "ferromagnetic helix" structure, for which the sums $r + \nu$ and $t + \tau$ may also be odd.

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