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ABSTRACT

For frequencies $m\alpha^2 \ll \omega \ll m\alpha$ and $m\alpha < \omega \ll m$ the dependent on the angular momentum amplitude of scattering of light on an atom is found. X-ray scattering on the crystal with magnetic order is considered. The scattering cross-section in the case of the helical spin structure for circularly polarized quanta depends not only on the period, but as well on the sign of the helix.

1. Introduction

In the work we consider the possibility to investigate a magnetic order by means of X-rays. Such experiments become quite feasible for synchrotron radiation (SR) since it possesses high intensity and is also elliptically polarized. The investigations of magnetic structures by means of SR may become an important addition to the neutron diffraction methods the latter being at present an effective origin of the information about the magnetic order in solids (see, e.g., Ref. [1]).

We obtain here the dependence of an X-ray scattering amplitude on the angular momentum of an atom or ion and consider scattering on a crystal with magnetic order. As an example we present the expression for the scattering cross-section on a crystal with a spin structure of the type "simple helix". The system of magnetic satellites around the main Bragg reflections is shown to give the information not only on the period of the helix, but due to the circular polarization of SR on the helix sign.

It should be noted that the possibility of the X-ray determination of the magnetic structure in collinear antiferromagnetics was not only discussed theoretically [2,3], but proven experimentally [4,5] for solids with incomplete d-shells. In the present work we point out and consider theoretically in detail other mechanisms of scattering dependent on angular momentum that are no less effective than those proposed in Refs. [2,3].

2. Asymptotics of dynamic polarizability of atom

The amplitude of elastic scattering of light on an isolated atom situated at the point $\vec{R}$ is in the dipole approximation [6]

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

Here $\vec{\varkappa} = \vec{k}' - \vec{k}$, $\vec{k}$ and $\vec{k}'$ are the wave vectors of incident and scattered photons, $\vec e$ and $\vec e\,'$ are the unit vectors of their polarizations. The units are used where $\hbar = c = 1$. The tensor of dynamic polarizability of atom in the state $|0\rangle$ is

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

where $\vec d$ is the dipole moment operator, $\omega_{n0} = E_n - E_0$, E_n and E_0 are the energies of the excited and ground states. If the state $|0\rangle$ is characterized by a total angular momentum $\vec J$, the tensor $\alpha_{ik}(\omega)$ can be written as

$$\alpha_{ik}(\omega) = \delta_{ik}\,\alpha_s(\omega) + i\varepsilon_{ikl}\,J_l\,\alpha_v(\omega) + Q_{ik}\,\alpha_t(\omega) \qquad (3)$$

where $Q_{ik} = J_iJ_k + J_kJ_i - \frac{2}{3}\;{}_{ik}J(J+1)$. The scalar $\alpha_s(\omega)$ vector $\alpha_v(\omega)$ and tensor $\alpha_t(\omega)$ polarizabilities due to the Hermiticity of the tensor $\alpha_{ik}(\omega)$ possess the following properties

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

The vector and tensor terms in (3) clearly contain the information about the orientation of the atomic total angular momentum.

We are interested in the scattering of the X-ray quanta (the frequency is $\omega \gg Ry = m\alpha^2/2$). Here m is electron mass, $\alpha = e^2 = 1/137$. We find the asymptotics of the polarizability at $\omega \to \infty$. The leading term of the series in $1/\omega$ for $\alpha_{ik}(\omega)$ is well-known

$$\alpha_{ik}(\omega) \simeq -\frac{Z'\alpha}{m\omega^2}\,\delta_{ik} \qquad (5)$$

It determines the scalar polarizability $\alpha_s(\omega)$. Here Z' is the number of external electrons for which the characteristic frequencies are much smaller than ω; $Z' \le Z$ where Z is the nuclear charge. The vector polarizability can appear in the next term of the series in $1/\omega$:

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

where $\hat H$ is an atomic Hamiltonian, $\vec r$ is an electron position

vector. The summation is carried over the atomic electrons. The expression (6) does not vanish only when we take into account in $\hat H$ the spin-orbit interaction, determining the asymptotics of the vector polarizability of atom. We shall show however that this contribution to $\alpha_v(\omega\to\infty)$ is not the leading one. The consistent calculation of relativistic corrections requires redetermination of the dipole moment operator in the expression (2).

The operator of the interaction of electron with electromagnetic field contains terms linear ($\hat H_1$) and quadratic ($\hat H_2$) in the field. If one takes into account the relativistic corrections, the operators are

$$\hat H_1 = -\frac{e}{2m}[\vec p,\vec A]_+ - \frac{e}{2m}\vec\sigma\vec H + \frac{e}{8m^2}\vec\sigma(\vec p\times\vec E - \vec E\times\vec p) \qquad (7)$$

$$\hat H_2 = \frac{\alpha}{2m}\vec A^2 - \frac{\alpha}{4m^2}\vec\sigma(\vec A\times\vec E) \qquad (8)$$

Here $[\cdot,\cdot]_+$ denote an anticommutator; $\vec A, \vec E, \vec H$ are vector potential, electric and magnetic field strengths respectively

$$\vec E = -\partial\vec A/\partial t,\quad \vec H = \mathrm{rot}\,\vec A,\quad \mathrm{div}\,\vec A = 0,\quad \vec A = \vec e\, e^{i\vec k\vec r - i\omega t}; \qquad (9)$$

$\vec p$ is the operator of the electron momentum, $\vec\sigma$ are the Pauli matrices. With the relativistic corrections taken into account the expression for the electrical dipole scattering amplitude is also reduced to (1), (2). If $\omega \ll m\alpha$, we have

$$-\frac{e}{2m}[\vec p,\vec A]_+ + \frac{e}{8m^2}\vec\sigma(\vec p\times\vec E - \vec E\times\vec p) = -ie[\hat H, \vec r\vec A + \frac{1}{4m}(\vec r\times\vec\sigma)\vec E]. \qquad (10)$$

Now the dipole moment operator in (1), (2) is

$$\vec d = e[\vec r + i\frac{\omega}{4m}(\vec r\times\vec\sigma)]. \qquad (11)$$

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

$$\alpha_{ik}(\omega\to\infty) \simeq i\frac{\alpha}{m^2\omega}\varepsilon_{ikl}S_l,\quad f^{E1,E1} \simeq i\frac{\alpha\omega}{m^2}(\vec e\,'^*\times\vec e)\vec S, \qquad (12)$$

where $\vec S$ is the total spin of atom. With (3), (12) taken into account the expression for the vector polarizability is

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

where $g = 1 + (\vec{J}\vec{S})/J(J+1)$ is Lande g-factor.

One can ascertain that just as the asymptotics (5) of scalar polarizability can be obtained without the transformation (10) directly from the first term in the expression (8), the asymptotics (13) corresponds to the second term in (8).

Find now the asymptotics of the tensor polarizability of atom, continuing the series in $1/\omega$ for (2)

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

here the one-electron Hamiltonian is $\hat{H} = \vec{p}^2/2m + U$. Trivial transformations allow one to present the second term in (14) as

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

The scalar part of this expression can be reduced to a small correction to the expression (5) for the scalar polarizability and is of no interest to us. For electrons with angular momentum $\ell \neq 0$ neglecting the term $\langle\Delta U\rangle$ in comparison with U'/r, we get for the tensor irreducible part of (15)

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

It follows from this expression that the tensor polarizability of a heavy atom with one external electron ($l \neq 0$) is

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

where $\vec{j}$ is the total angular momentum of the electron. The tensor polarizability of a heavy atom with some external electrons is found by summation of the expression (16) over the equivalent electrons of incomplete shell (or over equivalent holes). We are interested first of all in the ions Tb^{3+}, Ho^{3+}, Er^{3+}

where the f-shell contains more than half the greatest possible number of electrons for such a shell. Here in the ground state $J = L + S$, and the total spin S has as usually, according to the Hunds rule, its maximal value. The asymptotic expression for the tensor polarizability is reduced for these ions to

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

The analysis of fine structure of rare-earth ions and various numerical calculations (see, e.g., Ref. [10]) show that in the matrix element

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

the dimensionless factor ξ is close to 10. For frequency $m\alpha^2 \ll \omega \ll m\alpha$ which is considered in parts 2 and 3 $\alpha_t \ll \alpha_v$.

The fact that the tensor polarizability has the asymptotics $\sim\omega^{-4}$ was pointed out previously in Ref. [7].

The contribution of higher multipoles to the asymptotics of the scattering amplitude.

Since we take into account the relativistic correction to the electrical dipole scattering amplitude, it is quite natural to consider higher multipole amplitudes which arise in the same order in v/c. In particular we mean the magnetic scattering, discussed before in Refs. [2,3]. It is easy to show however that this order in v/c the electrical quadrupole scattering amplitude and interference amplitudes (E1,M2), (E1,E3) and (M1,E2) arise. The transformation of Hamiltonian (7) to

$$\begin{aligned}\hat{H}_1 = &-ie[\hat{H},(\vec{r}\vec{A}) + \frac{i}{2}(\vec{r}\vec{k})(\vec{r}\vec{A}) - \frac{1}{6}(\vec{r}\vec{k})^2(\vec{r}\vec{A})] - \\ &-\frac{e}{2m}\vec{H}(\vec{l}+\vec{\sigma}) - i\frac{e}{2m}(\vec{r}\vec{k})(\vec{H}\vec{\sigma}) - i\frac{e}{6m}[(\vec{r}\vec{k}),(\vec{H}\vec{l})]_+\end{aligned} \tag{20}$$

corresponds to the multipole expansion including E3 and M2. Relativistic corrections to the higher multipole amplitudes are unnecessary. Substituting (20) into the well-known expression for the light scattering amplitude

$$f = -\sum\left\{\langle \hat H_2\rangle - \sum_n\left[\frac{\langle 0|\hat H_1'|n\rangle\langle n|\hat H_1|0\rangle}{\omega_{n0}-\omega} + \frac{\langle 0|\hat H_1|n\rangle\langle n|\hat H_1'|0\rangle}{\omega_{n0}+\omega}\right]\right\} \qquad (21)$$

one can obtain the amplitudes corresponding to the contributions of the various multipoles. Here the radiation operator $\hat H_1'$ is connected with the absorption operator as

$$\hat H_1'(\vec e'^*, \omega, \vec k') = \hat H_1(\vec e, -\omega, -\vec k).$$

The electrical dipole (E1,E1)-amplitude we have considered in the previous part. The magnetic dipole (M1,M1)-amplitude is

$$f^{M1,M1} = \omega^2 (\vec n'\times\vec e'^*)_i (\vec n\times\vec e)_k\, \chi_{ik}(\omega) \qquad (22)$$

where $\vec n = \vec k/\omega$, $\vec n' = \vec k'/\omega$. The magnetic susceptibility tensor $\chi_{ik}(\omega)$ is

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

The primed summation symbol means as usually that the intermediate states do not include the ground one (in the expression (2) for $\alpha_{ik}(\omega)$ one should not bother about it specially due to the selection rules for E1 transitions). When expanding the tensor $\chi_{ik}(\omega)$ in $1/\omega$, already the first term does not vanish. It can be easily represented as

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

where the state $|0'\rangle$ can differ from $|0\rangle$ only by the projection M of the total angular momentum; the summation is carried out here over all M in the state $|0'\rangle$. From (24) one can obtain

$$\chi_{ik}(\omega\to\infty) \simeq i\,\varepsilon_{ikl}\,J_l\,\frac{\alpha(g-1)(g-2)}{4m^2\omega}. \qquad (25)$$

In the case of the ions interesting for us (g - 1) (g - 2) = = $-LS/J^2$. Note that the expression (25) determines the asymptotics of the vector magnetic susceptibility, the corresponding contribution to the scattering amplitude being proportional to ω :

$$f^{M1,M1} \simeq i\frac{\alpha\omega}{4m^2}\left[(\vec n'\times\vec e'^*)\times(\vec n\times\vec e)\right]\vec J(g-1)(g-2). \qquad (26)$$

Since the magnetic moment operator connects only the states with the same L and S and different J, the condition of the applicability of the expression (26) is extremely liberal: the frequency ω should only be much larger than the fine structure interval. On the other hand, in accordance with the condition of the multipole expansion for atoms we suppose here that $\omega \ll m\alpha$. In particular allows us to neglect the well-known diamagnetic contribution from $\hat H_2$ to the scattering amplitude. We make use of the condition $\omega \ll m\alpha$ systematically below in this part, restricting to the linear in ω terms in all amplitudes.

The asymptotic expression for the quadrupole (E2,E2)-amplitude we find easily

$$f^{E2,E2} \simeq -\frac{\alpha}{4}\omega\, e_i'^* e_k n_j' n_s \sum\langle[[\hat H, r_i r_j],[\hat H, r_k r_s]]\rangle. \qquad (27)$$

The summation is carried in this expression over all electrons. From (27) in the result of elementary transformations we obtain

$$f^{E2,E2} \simeq -i\frac{\alpha\omega}{4m^2}(g-2)\vec J\left[(\vec n\vec e'^*)(\vec n'\times\vec e) - (\vec n'\vec e)(\vec n\times\vec e'^*) +\right.$$
$$\left.+ (\vec n\vec n')(\vec e'^*\times\vec e) + (\vec e'^*\vec e)(\vec n'\times\vec n)\right]. \qquad (28)$$

Interference of the last two terms from (20) with the nonrelativistic E1 amplitude gives

$$\tilde f = i\frac{\varkappa\omega}{2m^2}g\vec J\left[(\vec n\vec e)(\vec n'\times\vec e'^*) - (\vec n\vec e'^*)(\vec n\times\vec e)\right] \qquad (29)$$

As regards the interference (E2,M1)- and (E1,E3)-amplitudes one can ascertain that in our approximation they vanish.

The total scattering amplitude depending on the angular momentum of atom or ion (the sum expressions (12), (26), (28) and (29)) for the frequencies $m\alpha^2 \ll \omega \ll m\alpha$ is

$$f = i\,\frac{\alpha\omega}{4m^2}\,(\vec{b}\vec{J}) \tag{30}$$

where

$$\begin{aligned}\vec{b} = {} & 4(g-1)(\vec{e}'^{*}\times\vec{e}) + (g-1)(g-2)(\vec{e}'^{*}\times\vec{n}')(\vec{e}\times\vec{n}) - \\ & -(g-2)\big[(\vec{n}\vec{e}'^{*})(\vec{n}'\times\vec{e}) - (\vec{n}'\vec{e})(\vec{n}\times\vec{e}'^{*}) + (\vec{n}\vec{n}')(\vec{e}'^{*}\times\vec{e}) + \\ & + (\vec{e}'^{*}\vec{e})(\vec{n}'\times\vec{n})\big] + 2g\big[(\vec{n}'\vec{e})(\vec{n}'\times\vec{e}'^{*}) - (\vec{n}\vec{e}'^{*})(\vec{n}\times\vec{e})\big].\end{aligned} \tag{31}$$

It should be noted that the second term in (31), corresponding to the magnetic amplitude $f^{M1,M1}$ is usual numerically shmal in comparison with others. The depended on angular momentum amplitude (30) is $\sim \omega/Z^2 m$ from the usual scalar amplitude, which from (1) and (5) is

$$f_1 = -\frac{Z\alpha}{m}(\vec{e}'^{*}\vec{e}). \tag{32}$$

The scattering amplitude for $\omega > m\alpha$

We have considered above the case $m\alpha^2 \ll \omega \ll m\alpha$. However the frequencies $m\alpha < \omega \ll m$ also are of great interest. Here the multipole expansion in the exploited above form cannot be applied, but the relativistic corrections still are too small and one can use the interaction operators (7) and (8).

Substitute the expressions for $\hat{H}_1$ and $\hat{H}_2$ into (21). The total scattering amplitude can be expressed as the sum of the five terms:

$$f_1 = -\frac{\alpha}{2m}\sum\langle \vec{A}^2\rangle, \tag{33}$$

$$f_2 = \frac{\alpha}{4m^2}\sum\langle \vec{\sigma}(\vec{A}\times\vec{E})\rangle, \tag{34}$$

$$f_3 = \frac{\alpha}{4m^2}\sum\Big[\frac{\langle 0|\vec{\sigma}\vec{H}'|n\rangle\langle n|\vec{\sigma}\vec{H}|0\rangle}{\omega_{n0}-\omega} + \frac{\langle 0|\vec{\sigma}\vec{H}|n\rangle\langle n|\vec{\sigma}\vec{H}'|0\rangle}{\omega_{n0}+\omega}\Big], \tag{35}$$

$$\begin{aligned} f_4 = \frac{\alpha}{2m^2}\sum\Big[& \frac{\langle 0|\vec{p}\vec{A}'|n\rangle\langle n|\vec{\sigma}\vec{H}|0\rangle}{\omega_{n0}-\omega} + \frac{\langle 0|\vec{\sigma}\vec{H}|n\rangle\langle n|\vec{p}\vec{A}'|0\rangle}{\omega_{n0}+\omega} + \\ & + \frac{\langle 0|\vec{\sigma}\vec{H}'|n\rangle\langle n|\vec{p}\vec{A}|0\rangle}{\omega_{n0}-\omega} + \frac{\langle 0|\vec{p}\vec{A}|n\rangle\langle n|\vec{\sigma}\vec{H}'|0\rangle}{\omega_{n0}+\omega}\Big], \end{aligned} \tag{36}$$

$$f_5 = \frac{\alpha}{m^2}\sum\Big[\frac{\langle 0|\vec{p}\vec{A}'|n\rangle\langle n|\vec{p}\vec{A}|0\rangle}{\omega_{n0}-\omega} + \frac{\langle 0|\vec{p}\vec{A}|n\rangle\langle n|\vec{p}\vec{A}'|0\rangle}{\omega_{n0}+\omega}\Big]. \tag{37}$$

At $m\alpha < \omega \ll m$ the expressions (33)-(37) are reduced to

$$f_1 = -\frac{\alpha}{m}(\vec{e}'^{*}\vec{e})\sum 4\pi(-i)^l\,Y^{*}_{l\tau}(\hat{\vec{\varkappa}})\langle j_l(\varkappa r)\rangle\langle Y_{l\tau}(\hat{\vec{r}})\rangle, \tag{38}$$

$$\begin{aligned} f &= f_2 + f_3 + f_4 = \\ &= -i\,\frac{\alpha\omega}{m^2}\sum 4\pi(-i)^l\,Y^{*}_{l\tau}(\hat{\vec{\varkappa}})\langle j_l(\varkappa r)\rangle\langle(\vec{a}\tfrac{\vec{\sigma}}{2})Y_{l\tau}(\hat{\vec{r}})\rangle, \end{aligned} \tag{39}$$

$$\begin{aligned} f_5 = {} & -i\,\frac{\alpha^2}{m\omega}\big[(\vec{e}'^{*}\times\vec{e})\times\vec{\varkappa}\big]\sum 4\pi(-i)^l\,Y^{*}_{l\tau}(\hat{\vec{\varkappa}})\times \\ & \times\Big[R_l(\varkappa)\langle Y_{l\tau}(\hat{\vec{r}})\hat{\vec{r}}\rangle - \frac{i}{2}S_l(\varkappa)\langle Y_{l\tau}(\hat{\vec{r}})(\hat{\vec{r}}\times\vec{l})\rangle\Big] \end{aligned} \tag{40}$$

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

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

$$S_t(x) = (m\alpha)^{-1} \langle j_t(xr)/r \rangle$$

are dimensicnless; $j_t(xr)$ is the spherical Bessel function;

$$\vec{a} = (\vec{n}'\times\vec{e}'^*)\times(\vec{n}\times\vec{e}) - (\vec{e}'^*\times\vec{e}) + \\ + (\vec{e}'^*\vec{n})(\vec{n}\times\vec{e}) - (\vec{e}\vec{n}')(\vec{n}'\times\vec{e}'^*) ; \tag{41}$$

the summation is carried over t , τ and all the atomic electrons. Obviously in the region $m\alpha \ll \omega \ll m$

$$f_s \sim \frac{\alpha^2}{m} \ll f \sim \frac{\alpha\omega}{m^2} \ll f_1 \sim \frac{\alpha}{m} , \tag{42}$$

therefore we neglect below the amplitude f_s. We note that both amplitudes f and f_1 depend on the orientation of the atomic angular momentum (except for the term in f_1 with t = 0). However, the dependence on the polarizations in f_1 is trivial $f_1 \sim (\vec{e}'^*\vec{e})$, therefore the smaller amplitude f is of more interest for the investigation of magnetic structure.

For the amplitude f_1 in the expression (38) it is necessary to calculate at $t \neq 0$ the sum of the one-electron matrix elements over the external shell

$$\sum \langle \ell m | Y_{t0}(\vec{r}) | \ell m \rangle , \tag{43}$$

m is the z-axis projection of the angular momentum $\vec{\ell}$ of an electron. Obviously in this expression t is even and limited ($t \leq 2\ell$). From the sum (43) it is convenient to pass to the equivalent operator in the space of the atomic angular momentum orientations. This operator should be by its transformation properties an irreducible tensor of the rank t (see, e.g., Ref. [9]) constructed from the angular momentum operators. We choose it in the form

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

Here $\{\vec{A}_a \otimes \vec{B}_a\}_\ell$ is the irreducible product of the rank b of the two operators of the rank a . Then we substitute the expression

$$\frac{1}{\sqrt{4\pi}} A^t_{JL\ell} \, T_{t\tau}(\vec{J}) \tag{45}$$

for the sum (43) in (38). Here the factors

$$A^t_{JL\ell} = \sqrt{4\pi} \, \frac{\langle J \| \vec{T}_t(\vec{L}) \| J \rangle \sum \langle \ell m | Y_{t0}(\vec{r}) | \ell m \rangle}{\langle J \| T_t(\vec{J}) \| J \rangle \langle L M_L | T_{t0}(\vec{L}) | L M_L \rangle} , \tag{46}$$

$$M_L = \sum m$$

one can calculate in the LS coupling scheme (see the Appendix). One can see that the factors $A^t_{JL\ell}$ are independent of the orientation of the vector $\vec{J}$, all the information about it containing in $T_{t\tau}(\vec{J})$. If we consider $\vec{J}$ as a classical vector directed along the unit vector $\vec{\mathcal{J}}$, then we have (see Ref. [9])

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

Quite analogously one can transform the amplitude f. After the expansion into the irreducible parts, with the Hund's rule (the total spin S has its maximal value) taken into account, we substitute the equivalent operator

$$\frac{1}{\sqrt{4\pi}} \sum_{\tau,\nu} (-1)^\nu a_{1-\nu} \, C^{\tau\,\tau+\nu}_{t\tau\,1\nu} \, A^{\tau t}_{JL\ell} \, T_{\tau\,\tau+\nu}(\vec{J}) \tag{48}$$

for the sum

$$\frac{1}{2} \sum \langle (\vec{a}\vec{\sigma}) Y_{t\tau}(\vec{r}) \rangle \tag{49}$$

in (39). Here the factor[1] (see Appendix)

1) One can ascertain that in case J = L + S $A^{rt}_{JL\ell} \neq 0$ only for $r = t \pm 1$.

$$A^{\tau t}_{JLe} = \sqrt{\frac{3\pi}{S(S+1)}} \cdot \frac{\langle J \| \{\vec{T}_t \otimes \vec{S}_1\}_\tau \| J\rangle \sum \langle \ell m | Y_{t0}(\hat{z}) | \ell m\rangle}{\langle J \| \vec{T}_\tau(\vec{J}) \| J\rangle \langle L M_L | T_{t0}(\vec{L}) | L M_L\rangle} \qquad (50)$$

is independent of the orientation of $\vec{J}$; $C^{c\gamma}_{a\alpha b\beta}$ are Clebsch-Gordan coefficients.

The Bragg scattering on a magnetic structure.

Below we shall be interested in the scattering amplitudes not of individual atoms or ions, but of a crystal with a certain magnetic structure, for example, a helical spin structure of the type "simple helix":

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

where $\vec{m}_1$ and $\vec{m}_2$ are the unit orthogonal vectors, $\vec{m} = \vec{m}_1 \times \vec{m}_2$; $\vec{g}$ is the wave vector of the helix, for the right helix $\vec{g}\vec{m} > 0$; index n numerates the unit cell, s numerates the ions of the unit cell. From the atomic amplitudes (30), (32), (38) and (39) one can pass to the scattering amplitudes of a crystal by summation over all ions with the weight factor $\exp(-i\vec{\varkappa}\vec{R}_{ns})$. In the case $\omega \ll m\alpha$ we obtain

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

$$F_2 = \sum_{n,s} f\, e^{-i\vec{\varkappa}\vec{R}_{ns}} =$$

$$= i\,\frac{\alpha\omega}{8m^2} N_1 \sum_{\vec{q}} J S(\vec{q})\,\vec{b}\,[\,\vec{m}_1 \delta^{(+1)} - i\,\vec{m}_2 \delta^{(-1)}\,], \qquad (53)$$

where $\vec{q}$ is the reciprocal lattice vector; N_τ is the number of unit cells contributing to the Bragg reflection with the momentum transfer $\vec{q} + \tau\vec{g}$;

$$\delta_{\vec{\varkappa},\vec{q}} = \begin{cases} 1 & \text{for } \vec{\varkappa} = \vec{q} \\ 0 & \text{for } \vec{\varkappa} \neq \vec{q}\,; \end{cases}$$

$\delta^{(\pm\tau)} = \delta_{\vec{\varkappa},\vec{q}+\tau\vec{g}} \pm \delta_{\vec{\varkappa},\vec{q}-\tau\vec{g}}$. For crystals with the hexagonal closed-packed lattice (just this lattice is characteristic for rare-earth metals of interest to us) the structure factor is

$$S(\vec{q}) = \sum_s e^{-i\vec{q}\vec{R}_s} = 2\cos(\vec{q}\vec{\rho}/2), \qquad (54)$$

where $\vec{\rho}$ is vector connecting two ions in the cell of hexagonal closed-packed structure.

In the case $\omega > m\alpha$ the dependence of the atomic amplitudes on the orientation of $\vec{J}$ is connected only with the tensor $T_{t\tau}(\vec{J})$. Directing the z-axis along the unit vector $\vec{m}$, we obtain with (47) and (51) taken into account

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

where for even $t + \tau$ $(t \neq 0)$

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

and for odd $t + \tau$ $\beta_{t\tau} = 0$. The scattering amplitudes for a crystal with magnetic structure of the type "simple helix" can be represented as

$$F_3 = -\frac{\alpha}{m}(\vec{e}'^{*}\vec{e}) \sum_{\vec{q},t,\tau} (-i)^t S(\vec{q}) N_\tau \langle j_t(\varkappa r)\rangle \times$$

$$\times\, J^t A^t_{JLe}\, \beta_{t\tau} \sqrt{4\pi}\, Y_{t\tau}(\hat{\varkappa})\, \delta_{\vec{\varkappa},\vec{q}+\tau\vec{g}}\,, \qquad (57)$$

$$F_4 = -i\,\frac{\alpha\omega}{m^2} \sum_{\substack{\vec{q},t\\ \tau,\nu}} (-i)^t S(\vec{q}) N_\nu \langle j_t(\varkappa r)\rangle \times$$

$$\times\, J^\tau A^{\tau t}_{JLe}\, \beta_{\tau\nu} \sqrt{4\pi}\, [\,\vec{a}\cdot \vec{Y}^{t*}_{\tau\nu}(\hat{\varkappa})\,]\, \delta_{\vec{\varkappa},\vec{q}+\nu\vec{g}}\,, \qquad (58)$$

where $\vec{Y}^{t}_{2\nu}(\vec{\varkappa})$ is a spheric vector (see Ref. [9]).

The indices t, τ $(t, |\tau| \leq 2\ell)$ in the expressions (57) and (58) are even the indices r, ν $(r, |\nu| \leq 2\ell + 1)$ are odd (see above). Due to it in the crystals where magnetic and crystallic structures are incommensurate ($\vec{q}$ and $\vec{g}$ have no common multiple) the amplitudes F_3 and F_4 do not interfere[2]. The amplitudes F_1 and F_2 do not interfere as well. However, the terms in (57) and (58) with equal τ or ν but noncoinciding other indices, can interfere among themselves. The terms with $|\tau| = t = 2\ell$ in (57) and with $|\nu| = r = 2\ell+1$ in (58) do not interfere with any other term.

Note that the expression (57) contains an isotropic term $(t = 0)$

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

with the summation over all electrons of ion. For $\varkappa r \ll 1$ this term competes with the amplitude F_1. The amplitude (59) for $\omega > m\alpha$ or (52) for $\omega \ll m\alpha$ determines the usual Bragg scattering with the momentum transfer $\vec{\varkappa} = \vec{q}$ (the Bragg reflex). In the region $\omega \ll m\alpha$ the pair of satellites with the relative intensity

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

appears at the Bragg reflexes. If $\omega > m\alpha$ the amplitudes (57) and (58) give the satellites the Bragg reflexes up to the order $2\ell + 1$ (for the rare-earth elements $l = 3$). For the satellites of the even order the intensity is given by the square of the amplitude (57), for odd by the square of (58). In particular it means the trivial dependence of the intensity of the even satellites on the polarization and the relative suppression of the odd satellites

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

2) This assertion for the structure of the type "ferromagnetic helix" is not valid. Here the sums $r + \nu$ and $t + \tau$ can be odd.

The intensity of the pair of satellites of the order $2\ell + 1$

As an example one can find the intensity of the satellites of the order $2l + 1$ at the Bragg reflexes in the scattering on the magnetic structure of the type "simple helix".

With the account for previous arguments the scattering cross-section is $(l = 3)$

$$|F_Y|^2 \simeq \frac{\alpha^2\omega^2}{m^4} N_7^2 \sum_{\vec{q}} 4\pi S^2(\vec{q}) \langle j_6(\varkappa r)\rangle^2 (A^{76}_{JL3})^2 J^{14} \times$$

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

$$+ a^{*}_{11} a_{11} \beta^2_{7-7} Y^{*}_{6-6}(\hat{\vec{\varkappa}}) Y_{6-6}(\hat{\vec{\varkappa}})\, \delta_{\vec{\varkappa},\vec{q}-7\vec{g}} \},$$

where the identities $C^{K+1,K+1}_{KK11} = C^{K+1,-K-1}_{K-K1-1} = 1$ are used. Taking also into account (56) and

$$|Y_{KK}(\hat{\vec{\varkappa}})|^2 = |Y_{K-K}(\hat{\vec{\varkappa}})|^2 =$$

$$= \frac{(2K+1)!!}{4\pi\,(2K)!!} \sin^{2K}\theta = \frac{(2K+1)!!}{4\pi\,(2K)!!} [1-(\hat{\vec{\varkappa}}\vec{m})^2]^K,$$

we obtain

$$|F_Y|^2 \simeq \frac{\alpha^2\omega^2}{m^4} N_7^2 \frac{15!!}{2^7\, 14!!} \sum_{\vec{q}} S^2(\vec{q}) \langle j_6(\varkappa r)\rangle^2 (A^{76}_{JL3})^2 J^{14} \times \qquad (61)$$

$$\times [1-(\hat{\vec{\varkappa}}\vec{m})^2]^6 (a^{*}_{1-1} a_{1-1} \delta_{\vec{\varkappa},\vec{q}+7\vec{g}} + a^{*}_{11} a_{11} \delta_{\vec{\varkappa},\vec{q}-7\vec{g}}).$$

Finally, passing to the Cartesian components of the vector $\vec{a}$ and calculating the factor A^{76}_{JL3} (see Appendix), we get in the case J = L + S

$$|F_4|^2 \simeq \frac{\alpha^2\omega^2}{m^4}\frac{3S}{(S+1)}\left[\frac{5!\,7!\,J^7(2J-7)!\,N_7}{3!\,3!\,2^7(2J)!(2S-1)!(6-2S)!}\right]^2 \sum_{\vec{q}} S^2(\vec{q})\times$$

$$\times\langle j_6(\varkappa z)\rangle^2\left[1-(\vec{\varkappa}\vec{m})^2\right]^6 a_i^* a_j \left[(\delta_{ij}-m_i m_j)\delta^{(+7)} - i\varepsilon_{ije} m_e \delta^{(-7)}\right] \tag{62}$$

The incident quanta are conveniently described by the polarization density matrix

$$\rho_{ik} = \frac{1}{2}(\delta_{ik}-n_i n_k) + \frac{\xi_1}{2}(e_{1i}e_{2k}+e_{2i}e_{1k}) - i\frac{\xi_2}{2}\varepsilon_{ike}n_e + \frac{\xi_3}{2}(e_{1i}e_{1k}-e_{2i}e_{2k}) \tag{63}$$

where ξ_1, ξ_2 and ξ_3 are the Stoke's parameters, $\vec{e}_1$ and $\vec{e}_2$ are the unit orthogonal vectors ($\vec{e}_1 \times \vec{e}_2 = \vec{n}$). We are not interested in the polarization of the scattered quanta. Substituting (63) into (62), we can obtain in the case $\vec{n} \parallel \vec{m}$

$$|F_4|^2 \simeq \frac{\alpha^2\omega^2}{m^4}\frac{3S}{(S+1)}\left[\frac{5!\,7!\,J^7(2J-7)!\,N_7}{3!\,3!\,2^{10}(2J)!(2S-1)!(6-2S)!}\right]^2 \sum_{\vec{q}} S^2(\vec{q})\times$$

$$\times\langle j_6(\varkappa z)\rangle^2(1+x)^6(1-x)\Big\{\left[(1+x)(2-x)+2\xi_1(\vec{e}_1\vec{n}')(\vec{e}_2\vec{n}')+\xi_3\left[(\vec{e}_1\vec{n}')^2-(\vec{e}_2\vec{n}')^2\right]\right]\delta^{(+7)} + \xi_2(1-x^2)\delta^{(-7)}\Big\} \tag{64}$$

where $x = (\vec{n}'\vec{n})$. From (64) it follows in particular that if the degree of the circular polarization of incident quanta ξ_2 is of the order of unity, then when the sign of the circular polarization or the sign of helix is changed the intensity of the given satellite changes also by a magnitude of the order of unity (note that the change of the sign of the helix corresponds to the change of the sign of $\vec{g}$). Thus, using the circular polarization of the synchrotron radiation one can define the sign of the magnetic helix for the single-domain pattern or the ratio of the volumes with domains of different signs.

The intensity of the pair of satellites of the order 21 + 1 can be compared with the intensity of the pair of the order 21. Taking into account (57), (56) and the Appendix in the case $\vec{n} \parallel \vec{m}$ we obtain

$$|F_3|^2 \simeq \frac{\alpha^2}{2m^2}\left[\frac{5!\,7!\,J^6(2J-6)!\,N_6}{3!\,3!\,2^9(2J)!(2S-1)!(6-2S)!}\right]^2 \sum_{\vec{q}} S^2(\vec{q})\times \langle j_6(\varkappa z)\rangle^2(1+x)^6(1+x^2)\delta^{(+6)} \tag{65}$$

for the part independent of the polarization. In accordance with the estimate made above

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

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__Appendix__

For the factors A^{t}_{JLl} and A^{rt}_{JLl} (see (46) and (50)) used in the our work one can obtain

$$A^{t}_{JLl} = (-1)^{J+L+S-t}\, 2^{t}(2J+1)\begin{Bmatrix} L & S & J \\ J & t & L \end{Bmatrix}\left[\frac{(2L+1)(2J-t)!\,(2t+1)!!}{t!\,(2J+t+1)!}\right]^{1/2} B^{t}_{Ll}\,, \tag{A.1}$$

$$A^{rt}_{JLl} = 2^{r-1}(2J+1)\begin{Bmatrix} t & 1 & r \\ L & S & J \\ L & S & J \end{Bmatrix}\left[\frac{3(2S+1)(2L+1)(2t+1)(2J-r)!\,(2r+1)!!}{r!\,(2J+r+1)!}\right]^{1/2} B^{t}_{Ll} \tag{A.2}$$

where, according to the Hund's rule,

$$B^{t}_{Ll} = C^{l0}_{l0t0}\left(\sum_{m=l-2S+1}^{l} C^{lm}_{lmt0}\right) \Big/ C^{LL}_{LLt0} \tag{A.3}$$

The explicit expressions for $6j$-symbol in (A.1) or $9j$-symbol in (A.2) one can simplify in the case J = L + S of interest to us. Since the $9j$-symbol differs from zero only for $r = t\pm1$, we get

$$A^{t}_{JLl} = 2^{t}\frac{(2L)!\,(2J-t)!}{(2J)!}\left[\frac{(2L+1)(2t+1)!!}{t!\,(2L-t)!\,(2L+t+1)!}\right]^{1/2} B^{t}_{Ll}\,, \tag{A.4}$$

$$A^{t+1\,t}_{JLl} = 2^{t}\frac{(2L)!\,(2J-t-1)!}{(2J)!}\left[\frac{3S(2L+1)(2t+1)!!}{(S+1)\,t!\,(2L-t)!\,(2L+t+1)!}\right]^{1/2} B^{t}_{Ll} \tag{A.5}$$

$$A^{t-1\,t}_{JLl} = -2^{t-2}\frac{(2L)!\,(2J-t+1)!}{(2J)!}\left[\frac{3S(2L+1)\,t\,(2t-3)!!}{(S+1)(t-1)!\,(2L-t)!\,(2L+t+1)!}\right]^{1/2} B^{t}_{Ll}. \tag{A.6}$$

The expression (A.3) for B^{t}_{Ll} simplifies mostly in the case $t = 2l$

$$B^{2l}_{Ll} = (-1)^{2S+l+1}\frac{(2l)!\,(2l)!\,(2l+1)!\,(2l-1)!}{l!\,l!\,(4l+1)!\,(2S-1)!\,(2l-2S)!}\left[\frac{(2L+2l+1)!\,(2L-2l)!}{(2L)!\,(2L+1)!}\right]^{1/2}, \tag{A.7}$$

For $t = 2l$ the factors (A.4) - (A.6) are

$$A^{2l}_{JLl} = (-1)^{2S+l+1}\frac{2^{l}(2J-2l)!\,(2l)!\,(2l+1)!\,(2l-1)!}{(2J)!\,l!\,l!\,(2S-1)!\,(2l-2S)!\,\sqrt{(4l+1)!}}\,, \tag{A.8}$$

$$A^{2l+1,2l}_{JLl} = (-1)^{2S+l+1}\frac{2^{l}(2J-2l-1)!\,(2l)!\,(2l+1)!\,(2l-1)!}{(2J)!\,l!\,l!\,(2S-1)!\,(2l-2S)!}\left[\frac{3S}{(S+1)(4l+1)!}\right]^{1/2}, \tag{A.9}$$

$$A^{2l-1,2l}_{JLl} = \frac{(-1)^{2S+l}\,2^{l-2}(2J-2l+1)!\,(2l)!\,(2l)!\,(2l+1)!\,\sqrt{3S}}{(2J)!\,l!\,l!\,(2S-1)!\,(2l-2S)!\,\sqrt{(S+1)(4l+1)!\,(4l+1)(4l-1)}}\,. \tag{A.10}$$

References

1. Izyumov Ju.A., Najsh B.E., Ozerov R.P. Neutrons and Solids v. 2. Neutronography of Magnetics (in Russian), Moscow, Atomizdat, 1981.
2. Platzman P.M., Tzoar N. Phys. Rev. B., 1970, 2, 3556.
3. Babushkin R.A. Dynamic Theory of X-Ray Scattering on Antiferromagnetics (in Russian), Leningrad, 1979.
4. De Bergevin F., Brunel M. Phys. Lett., 1972, A39, 141.
5. Brunel M., De Bergevin F. Proc. Int. Conf. Magnet. (MKM-73), v. 2, Moscow, 1974.
6. Berestetskii V.B., Lifshitz E.M., Pitayevskii L.P. Quantum Electrodynamics (in Russian), Moscow, Nauka, 1980.
7. Manakov N.L., Ovsyannikov V.D. Zh. Eksp. Teor. Fiz., 1978, 75, 803.
8. Taylor K.N.R., Darby M.I. Physics of Rare-Earth Solids. Chairman and Hall ltd, London, 1972.
9. Varshalovich D.A., Moskalev A.N., Khersonskii V.K. Quantum Theory of Angular Momentum (in Russian), Leningrad, Nauka, 1975.

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К ВОЗМОЖНОСТИ РЕНТГЕНОВСКОГО ИССЛЕДОВАНИЯ МАГНИТНОЙ СТРУКТУРЫ КРИСТАЛЛОВ

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