X-ray Resonant Exchange Scattering (XRES)

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Derivation of the scattering amplitude for XRES as first given by: J. P. Hannon, G. T. Trammell, M. Blume and D. Gibbs, Phys. Rev. Lett. 61,1245 (1988); 62, 2644(E) (1989)

Starting from the non-relativistic Hamiltonian for electrons in an electromagnetic field

$$H = \sum_i \frac{1}{2m}\left(\mathbf{p}_i - \frac{e}{c}\mathbf{A}(\mathbf{r}_i)\right)^2 \tag{1}$$

one can show, using Fermi's golden rule, that the elastic scattering amplitude f is given by [1] :

$$f = r_0 \left\langle a \left| \sum_i e^{i\mathbf{K}\mathbf{r}_i} \right| a \right\rangle \boldsymbol{\epsilon}^*(\mathbf{k}').\boldsymbol{\epsilon}(\mathbf{k}) + \frac{r_0}{m}\sum_c \sum_{i,j}\left[\frac{\left\langle a \left| \boldsymbol{\epsilon}^*(\mathbf{k}')e^{i\mathbf{k}'\mathbf{r}_i}\mathbf{p}_i \right| c \right\rangle \left\langle c \left| \boldsymbol{\epsilon}(\mathbf{k})e^{i\mathbf{k}\mathbf{r}_j}\mathbf{p}_j \right| a \right\rangle}{E_a - E_c + \hbar\omega_k - \frac{i\Gamma_c}{2}}\right] \tag{2}$$

Here $\boldsymbol{\epsilon}$ ($\boldsymbol{\epsilon}'$) and $\mathbf{k}$ ($\mathbf{k}'$) are the unit polarization vector resp. wave vector of the incident (outgoing) photon, and $\mathbf{K} = \mathbf{k} - \mathbf{k}'$. $r_0 = \frac{e^2}{mc^2} = 2.82\ 10^{-13} cm$. $|\, a\rangle$ is the ground state of the system and $|\, c\rangle$ some intermediate state with energy E_c and total level width Γ_c. The first term gives the usual expression for Bragg scattering; the second term accounts for anomalous dispersion effects and, due to the spin-orbit split empty states above the Fermi level, also to magnetic effects. This has been shown by Hannon et al. [2] who derive an expression for the resonant terms of the scattering amplitude in terms of the partial width for EL radiative decay from $|\, c\rangle$ to $|\, a\rangle$. Since nowhere in literature a derivation of their expression (which is by no means straightforward) is given, we will show extensively how to repeat their calculation. We first write $\boldsymbol{\epsilon}(\mathbf{k})e^{i\mathbf{k}\mathbf{r}_j}$ as [3] :

$$\boldsymbol{\epsilon}(\mathbf{k})e^{i\mathbf{k}\mathbf{r}_j} = 4\pi \sum_{J=0}^{\infty}\sum_{M=-J}^{J}\left[\sum_{L=J-1}^{J+1} i^L j_L(kr_j)\left\{\boldsymbol{\epsilon}(\mathbf{k})\mathbf{Y}_{JM}^{L*}(\hat{\mathbf{k}})\right\}\mathbf{Y}_{JM}^{L}(\hat{\mathbf{r}}_j)\right] \tag{3}$$

where $\mathbf{Y}_{JM}^{L}$ are vector spherical harmonics and j_L are spherical Bessel functions. We introduce the vector spherical harmonics $\mathbf{Y}_{JM}^{(\lambda)}$ ($\lambda = -1, 0, 1$) as [4]:

$$\mathbf{Y}_{JM}^{(1)}(\hat{r}_j) \quad = \quad \frac{1}{\sqrt{J(J+1)}}\nabla_\Omega Y_{JM}(\hat{r}_j)$$

$$\begin{aligned}
\mathbf{Y}^{(0)}_{JM}(\hat{r}_j) &= \frac{-i}{\sqrt{J(J+1)}}(\mathbf{n}\times\nabla_\Omega)Y_{JM}(\hat{r}_j) \\
\mathbf{Y}^{(-1)}_{JM}(\hat{r}_j) &= \mathbf{n}Y_{JM}(\hat{r}_j)
\end{aligned} \tag{4}$$

Here, ∇_Ω denotes the angular part of the ∇ operator and $\mathbf{n} = \mathbf{r}/r$. Using the relations between the $\mathbf{Y}^{(\lambda)}_{JM}$ and $\mathbf{Y}^{L}_{JM}$ vector spherical harmonics [5], one can write :

$$\begin{aligned}
\boldsymbol{\epsilon}(\mathbf{k})e^{i\mathbf{k}\mathbf{r}_j} = 4\pi\sum_{JM}\Big[& i^{J+1}\left\{\boldsymbol{\epsilon}(\mathbf{k})\mathbf{Y}^{(1)*}_{JM}(\hat{\mathbf{k}})\right\}\mathbf{A}_{JM}(\mathbf{r}_j,e)+ \\
& i^{J}\left\{\boldsymbol{\epsilon}(\mathbf{k})\mathbf{Y}^{(0)*}_{JM}(\hat{\mathbf{k}})\right\}\mathbf{A}_{JM}(\mathbf{r}_j,m)+i^{J-1}\left\{\boldsymbol{\epsilon}(\mathbf{k})\mathbf{Y}^{(-1)*}_{JM}(\hat{\mathbf{k}})\right\}\mathbf{A}_{JM}(\mathbf{r}_j,l)\Big]
\end{aligned} \tag{5}$$

with :

$$\begin{aligned}
\mathbf{A}_{JM}(\mathbf{r},e) &= \sqrt{\frac{J}{2J+1}}j_{J+1}(kr)\mathbf{Y}^{J+1}_{JM}(\hat{r}) - \sqrt{\frac{J+1}{2J+1}}j_{J-1}(kr)\mathbf{Y}^{J-1}_{JM}(\hat{r}) \\
\mathbf{A}_{JM}(\mathbf{r},m) &= j_J(kr)\mathbf{Y}^{J}_{JM}(\hat{r}) \\
\mathbf{A}_{JM}(\mathbf{r},l) &= \sqrt{\frac{J+1}{2J+1}}j_{J+1}(kr)\mathbf{Y}^{J+1}_{JM}(\hat{r}) + \sqrt{\frac{J}{2J+1}}j_{J-1}(kr)\mathbf{Y}^{J-1}_{JM}(\hat{r})
\end{aligned} \tag{6}$$

The quantities $\mathbf{A}_{JM}(\mathbf{r},a)$ ($a = e, m$ or l) are called the multipole potentials. They are all solutions of the vector Helmholtz equations (see e.g. [6, 7]). Since :

$$\nabla\mathbf{A}_{JM}(\mathbf{r},l) \neq 0 \tag{7}$$

this potential does not satisfy the condition of transverseness (hence the label l for *longitudinal*) so it will not be used in describing the electromagnetic field in the transverse gauge which we will use. The fields associated with the multipole potentials are defined as :

$$\begin{aligned}
\mathbf{E}_{JM}(\mathbf{r},a) &= ik\mathbf{A}_{JM}(\mathbf{r},a) \\
\mathbf{H}_{JM}(\mathbf{r},a) &= \nabla\times\mathbf{A}_{JM}(\mathbf{r},a), \qquad (a = e \text{ or } m)
\end{aligned} \tag{8}$$

It follows directly from Maxwell's equations that in the near (or static) zone, where $kr \to 0$, and the Bessel functions may be replaced by their leading terms in the expansion [8]:

$$j_J(kr) \to j^0_J(kr) = \frac{(kr)^J}{(2J+1)!!} \tag{9}$$

one has :

$$\begin{aligned}
H_{JM}(\mathbf{r},e) &\sim (kr)H_{JM}(\mathbf{r},m) \ll H_{JM}(\mathbf{r},m) \\
E_{JM}(\mathbf{r},m) &\sim (kr)E_{JM}(\mathbf{r},e) \ll E_{JM}(\mathbf{r},e)
\end{aligned} \tag{10}$$

So the labels e and m of the multipole vector potentials are chosen such that in the near zone the e-component of the electric field dominates over its m-component, and the m-component of the magnetic field dominates over its e-component. Since we are only interested here in electric multipole contributions we can write from (5):

$$\boldsymbol{\epsilon}(\mathbf{k})e^{i\mathbf{k}\mathbf{r}_j} = 4\pi \sum_{JM} i^{J+1} \left\{\boldsymbol{\epsilon}(\mathbf{k})\mathbf{Y}^{(1)*}_{JM}(\hat{\mathbf{k}})\right\} \mathbf{A}_{JM}(\mathbf{r}_j, e) \tag{11}$$

Now in the near zone, we can use expansion (9) for the Bessel functions, so :

$$\begin{aligned}
\mathbf{A}_{JM}(\mathbf{r}, e) &\rightarrow -\sqrt{\frac{J}{2J+1}}\frac{(kr)^{J-1}}{(2(J-1)+1)!!}\mathbf{Y}^{J-1}_{JM}(\hat{r}) \\
&\rightarrow -\sqrt{\frac{J+1}{J}}\mathbf{A}_{JM}(\mathbf{r}, l) \\
&\rightarrow -\sqrt{\frac{J+1}{J}}\frac{1}{k}\nabla\left(j^0_J(kr)Y_{JM}(\hat{r})\right)
\end{aligned} \tag{12}$$

We thus find :

$$\left\langle c \left| \boldsymbol{\epsilon}(\mathbf{k})e^{i\mathbf{k}\mathbf{r}_j}\mathbf{p}_j \right| a\right\rangle = -4\pi \sum_{JM} i^{J+1} \left\{\boldsymbol{\epsilon}(\mathbf{k})\mathbf{Y}^{(1)*}_{JM}(\hat{\mathbf{k}})\right\} \sqrt{\frac{J+1}{J}}\frac{1}{k}\left\langle c \left| \nabla\left(j^0_J(kr_j)Y_{JM}(\hat{r}_j)\right)\mathbf{p}_j \right| a\right\rangle \tag{13}$$

Using partial integration and the equation of continuity it is easy to show that :

$$\left\langle c \left| \nabla\left(j^0_J(kr_j)Y_{JM}(\hat{r}_j)\right)\mathbf{p}_j \right| a\right\rangle = im\omega \left\langle c \left| j^0_J(kr_j)Y_{JM}(\hat{r}_j) \right| a\right\rangle \tag{14}$$

This way the contribution of a 2^J-pole resonance to the elastic scattering amplitude (2) can be written (assuming $|\mathbf{k}'| = |\mathbf{k}| = k$) :

$$\begin{aligned}
f^{EJ}_{XRES} = 16\pi^2 e^2 \sum_c \sum_M \frac{J+1}{J} &\left\{\boldsymbol{\epsilon}'^*(\mathbf{k}')\mathbf{Y}^{(1)}_{JM}(\hat{\mathbf{k}}')\mathbf{Y}^{(1)*}_{JM}(\hat{\mathbf{k}})\boldsymbol{\epsilon}(\mathbf{k})\right\} \\
&\frac{\langle a|\sum_i j^0_J(kr_i)Y^*_{JM}(\hat{r}_i)|c\rangle\langle c|\sum_i j^0_J(kr_j)Y_{JM}(\hat{r}_j)|a\rangle}{E_a - E_c + \hbar\omega_k - \frac{i\Gamma_c}{2}}
\end{aligned} \tag{15}$$

This is the $T = 0$ expression for the x-ray resonant exchange scattering amplitude as it was derived in [2]. Using (9) we get for the dipole (J=1) contribution :

$$\begin{aligned}
f^{E1}_{XRES} &= r_0\frac{32\pi^2 mc^2k^2}{9} \sum_{M=-1}^{1} \left\{\boldsymbol{\epsilon}'^*(\mathbf{k}')\mathbf{Y}^{(1)}_{1M}(\hat{\mathbf{k}}')\mathbf{Y}^{(1)*}_{1M}(\hat{\mathbf{k}})\boldsymbol{\epsilon}(\mathbf{k})\right\} \sum_c \frac{\left|\left\langle a\left|\sum_j r_j Y^*_{1M}(\hat{r}_j)\right|c\right\rangle\right|^2}{E_a - E_c + \hbar\omega_k - \frac{i\Gamma_c}{2}} \\
&= 2\lambda \sum_{M=-1}^{1} \left\{\boldsymbol{\epsilon}'^*(\mathbf{k}')\mathbf{Y}^{(1)}_{1M}(\hat{\mathbf{k}}')\mathbf{Y}^{(1)*}_{1M}(\hat{\mathbf{k}})\boldsymbol{\epsilon}(\mathbf{k})\right\} F^{(e)}_{1M}
\end{aligned} \tag{16}$$

with :

$$F^{(e)}_{1M} = \frac{16\pi^2 e^2 k^2}{9\lambda} \sum_c \frac{\left|\left\langle a\left|\sum_j r_j Y^*_{1M}(\hat{r}_j)\right|c\right\rangle\right|^2}{E_a - E_c + \hbar\omega_k - \frac{i\Gamma_c}{2}} \tag{17}$$

Polarization dependence

The polarization dependence of the XRES scattering amplitude is expressed by :

$$\left\{\boldsymbol{\epsilon}'^{*}(\mathbf{k}')\mathbf{Y}^{(1)}_{JM}(\hat{\mathbf{k}}')\mathbf{Y}^{(1)*}_{JM}(\hat{\mathbf{k}})\boldsymbol{\epsilon}(\mathbf{k})\right\} \tag{18}$$

We will, for the case of dipole transitions, simplify this expression. We start by proving the following identity [9] :

$$\boldsymbol{\epsilon}(\mathbf{k})\mathbf{Y}^{(1)}_{JM}(\hat{\mathbf{k}}) = \sqrt{\frac{4\pi(2J+1)}{3(J+1)}}\sum_{\mu} C^{JM}_{J-1\ M-\mu\ 1\mu}Y_{J-1\ M-\mu}(\hat{\mathbf{k}})Y_{1\mu}(\hat{\boldsymbol{\epsilon}}) \tag{19}$$

We first expand $\mathbf{Y}^{(1)}_{JM}(\hat{\mathbf{k}})$ in terms of the spherical covariant basis vectors $\mathbf{e}_\mu$ ($\mu = \pm 1, 0$) [10]:

$$\begin{aligned}
\mathbf{Y}^{(1)}_{JM}(\hat{\mathbf{k}}) &= \textstyle\sum_\mu \left[\ Y^{(1)}_{JM}(\hat{\mathbf{k}})\right]^\mu \mathbf{e}_\mu \\
&= \textstyle\sum_\mu \Big[\ \sqrt{\tfrac{J+1}{2J+1}}C^{JM}_{J-1\ M-\mu\ 1\mu}Y_{J-1\ M-\mu}(\hat{\mathbf{k}}) \quad + \\
&\qquad \sqrt{\tfrac{J}{2J+1}}C^{JM}_{J+1\ M-\mu\ 1\mu}Y_{J+1\ M-\mu}(\hat{\mathbf{k}}) \ \Big]\mathbf{e}_\mu
\end{aligned} \tag{20}$$

where $C^{JM}_{j_1m_1j_2m_2}$ are the Clebsch-Gordan coefficients. Then we use the following expansion of the product of two spherical harmonics [11]:

$$Y_{j_1m_1}(\theta_k,\phi_k)Y_{j_2m_2}(\theta_k,\phi_k) = \sum_{JM}\sqrt{\frac{(2j_1+1)(2j_2+1)}{4\pi(2J+1)}}C^{J0}_{j_10\ j_20}C^{JM}_{j_1m_1j_2m_2}Y_{JM}(\theta_k,\phi_k) \tag{21}$$

with $\hat{\mathbf{k}} = (\theta_k, \phi_k)$. This expression together with the fact that we can write the components of any vector $\mathbf{k}$ as [12] :

$$k_\mu = \sqrt{\frac{4\pi}{3}}\,|\mathbf{k}|\,Y_{1\mu}(\theta_k,\phi_k) \tag{22}$$

gives the following recursion relations for the scalar spherical harmonics :

$$\begin{aligned}
\hat{k}_{-1}Y_{JM}(\hat{\mathbf{k}}) &= \sin\theta_k e^{-i\phi_k}Y_{JM}(\hat{\mathbf{k}}) \\
&= \sqrt{\frac{(J-M+1)(J-M+2)}{(2J+1)(2J+3)}}Y_{J+1\ M-1}(\hat{\mathbf{k}}) - \\
&\qquad \sqrt{\frac{(J+M-1)(J+M)}{(2J-1)(2J+1)}}Y_{J-1\ M}(\hat{\mathbf{k}}) \\
\hat{k}_0 Y_{JM}(\hat{\mathbf{k}}) &= \cos\theta_k Y_{JM}(\hat{\mathbf{k}})
\end{aligned}$$

$$
\begin{aligned}
&= \sqrt{\frac{(J-M+1)(J+M+1)}{(2J+1)(2J+3)}} Y_{J+1\ M}(\hat{\mathbf{k}}) + \\
&\quad \sqrt{\frac{(J-M)(J+M)}{(2J-1)(2J+1)}} Y_{J-1\ M}(\hat{\mathbf{k}}) \\
\hat{k}_1 Y_{JM}(\hat{\mathbf{k}}) &= \sin\theta_k e^{i\phi_k} Y_{JM}(\hat{\mathbf{k}}) \\
&= -\sqrt{\frac{(J+M+1)(J+M+2)}{(2J+1)(2J+3)}} Y_{J+1\ M}(\hat{\mathbf{k}}) + \\
&\quad \sqrt{\frac{(J-M-1)(J-M)}{(2J-1)(2J+1)}} Y_{J-1\ M}(\hat{\mathbf{k}}) \qquad (23)
\end{aligned}
$$

Inserting (23) into (20) gives :

$$
\mathbf{Y}^{(1)}_{JM}(\hat{\mathbf{k}}) = \sqrt{\frac{2J+1}{J+1}} \sum_{\mu} C^{JM}_{J-1\ M-\mu\ 1\mu} Y_{J-1\ M-\mu}(\hat{\mathbf{k}}) \mathbf{e}_\mu - \sqrt{\frac{J}{J+1}} \hat{\mathbf{k}} Y_{JM}(\hat{\mathbf{k}}) \qquad (24)
$$

Since we are dealing with transverse waves we have $\boldsymbol{\epsilon}(\mathbf{k}).\hat{\mathbf{k}} = 0$. Then using the expansion (22) for $\boldsymbol{\epsilon}(\mathbf{k})$ we arrive at (19).
For dipole transitions (J=1) expression (19) becomes :

$$
\boldsymbol{\epsilon}^{'*}(\mathbf{k}^{'}) \mathbf{Y}^{(1)}_{1M}(\hat{\mathbf{k}}) = \sqrt{\frac{3}{8\pi}} \boldsymbol{\epsilon}^{'*}(\mathbf{k}^{'}) \mathbf{e}_M \qquad (25)
$$

Then using :

$$
\mathbf{Y}^{(\lambda)*}_{JM} = (-)^{M+\lambda+1} \mathbf{Y}^{\lambda}_{J\ -M} \qquad (26)
$$

we find :

$$
\left(\boldsymbol{\epsilon}^{'*}(\mathbf{k}^{'}) \mathbf{Y}^{(1)}_{1M}(\hat{\mathbf{k}}^{'})\right) \left(\mathbf{Y}^{(1)*}_{1M}(\hat{\mathbf{k}}) \boldsymbol{\epsilon}(\mathbf{k})\right) = -\frac{3}{8\pi} \left(\boldsymbol{\epsilon}^{'*}(\mathbf{k}^{'}) \mathbf{e}_M\right) \left(\mathbf{e}_{-M} \boldsymbol{\epsilon}(\mathbf{k})\right) \qquad (27)
$$

Now we will relate expression (27) for all values of M to only one of the spherical basis vectors, say $\mathbf{e}_0$, which will be taken in the direction of the local quantization axis of the magnetic ion. Using the relations :

$$
\begin{aligned}
\mathbf{e}_\mu \times \mathbf{e}_\nu &= i\sqrt{2} C^{1\lambda}_{1\mu 1\nu} \mathbf{e}_\lambda \\
\mathbf{e}_\mu . \mathbf{e}^*_\nu &= \delta_{\mu\nu} \qquad (28)
\end{aligned}
$$

and the properties of the cross-product, it is straightforward to show that one arrives at [13]:

$$
\begin{aligned}
\left(\boldsymbol{\epsilon}^{'*}(\mathbf{k}^{'}) \mathbf{Y}^{(1)}_{10}(\hat{\mathbf{k}}^{'})\right) \left(\mathbf{Y}^{(1)*}_{10}(\hat{\mathbf{k}}) \boldsymbol{\epsilon}(\mathbf{k})\right) &= \frac{3}{8\pi} (\boldsymbol{\epsilon}^{'*} \mathbf{e}_0)(\mathbf{e}_{-0} \boldsymbol{\epsilon}) \\
\left(\boldsymbol{\epsilon}^{'*}(\mathbf{k}^{'}) \mathbf{Y}^{(1)}_{11}(\hat{\mathbf{k}}^{'})\right) \left(\mathbf{Y}^{(1)*}_{11}(\hat{\mathbf{k}}) \boldsymbol{\epsilon}(\mathbf{k})\right) &= \frac{3}{16\pi} \left[(\boldsymbol{\epsilon}^{'*}.\boldsymbol{\epsilon}) - i(\boldsymbol{\epsilon}^{'*} \times \boldsymbol{\epsilon}) \mathbf{e}_0 - (\boldsymbol{\epsilon}^{'*} \mathbf{e}_0)(\mathbf{e}_0 \boldsymbol{\epsilon})\right] \\
\left(\boldsymbol{\epsilon}^{'*}(\mathbf{k}^{'}) \mathbf{Y}^{(1)}_{1-1}(\hat{\mathbf{k}}^{'})\right) \left(\mathbf{Y}^{(1)*}_{1-1}(\hat{\mathbf{k}}) \boldsymbol{\epsilon}(\mathbf{k})\right) &= \frac{3}{16\pi} \left[(\boldsymbol{\epsilon}^{'*}.\boldsymbol{\epsilon}) + i(\boldsymbol{\epsilon}^{'*} \times \boldsymbol{\epsilon}) \mathbf{e}_0 - (\boldsymbol{\epsilon}^{'*} \mathbf{e}_0)(\mathbf{e}_0 \boldsymbol{\epsilon})\right] \qquad (29)
\end{aligned}
$$

Finally, combining (16) and (29) we find :

$$f^{E1}_{XRES} = \frac{3\lambda}{8\pi}\Big[(\boldsymbol{\epsilon}'^{*}.\boldsymbol{\epsilon})(F^{(e)}_{11} + F^{(e)}_{1-1}) - i(\boldsymbol{\epsilon}'^{*} \times \boldsymbol{\epsilon})(F^{(e)}_{11} - F^{(e)}_{1-1}) + (\boldsymbol{\epsilon}'^{*}\mathbf{e}_0)(\mathbf{e}_0\boldsymbol{\epsilon})(2F^{(e)}_{10} - F^{(e)}_{11} - F^{(e)}_{1-1})\Big] \tag{30}$$

This proves equation (3) in [2]

References

[1] M. Blume, J. Appl. Phys. **57** (1985) 3615.

[2] J. P. Hannon, G. T. Trammell, M. Blume and D. Gibbs, Phys. Rev. Lett. **61** (1988) 1245; **62** (1989) 2644(E).

[3] D.A.Varshalovich, A.N.Moskalev and V.K.Khersonskii, *Quantum Theory of Angular Momentum*, World Scientific, Singapore, 1988, p228, section 7.3.13.

[4] $\mathbf{Y}^{(1)}_{JM}(\hat{(r)})$ and $\mathbf{Y}^{(0)}_{JM}(\hat{(r)})$ are transverse and $\mathbf{Y}^{(-1)}_{JM}(\hat{(r)})$ is longitudinal with respect to $\mathbf{n}$. The three vectors $\mathbf{Y}^{(\lambda)}_{JM}$ with different λ are mutually orthogonal.

[5] Reference [3], p210, section 7.3.1.

[6] S. DeBenedetti, *Nuclear Interactions*, John Wiley and Sons Inc., New York, 1964, Section 4.13.

[7] J.M.Eisenberg and W. Greiner, *Nuclear Theory, Volume 2 : Excitation Mechanisms of the Nucleus*, North-Holland Publishing company, Amsterdam 1970, Chapter 3.

[8] $(2J+1)!!$ is defined as $(2J+1).(2J-1).(2J-3)...1$

[9] This expression for $\boldsymbol{\epsilon}(\mathbf{k})\mathbf{Y}^{(1)}_{JM}(\hat{\mathbf{k}})$ is also used in [2].

[10] Reference [3], p212, section 7.3.2

[11] Reference [3], p144, section 5.6.2.

[12] Reference [3], p13, section 1.2.

[13] These identities are also given in [2].