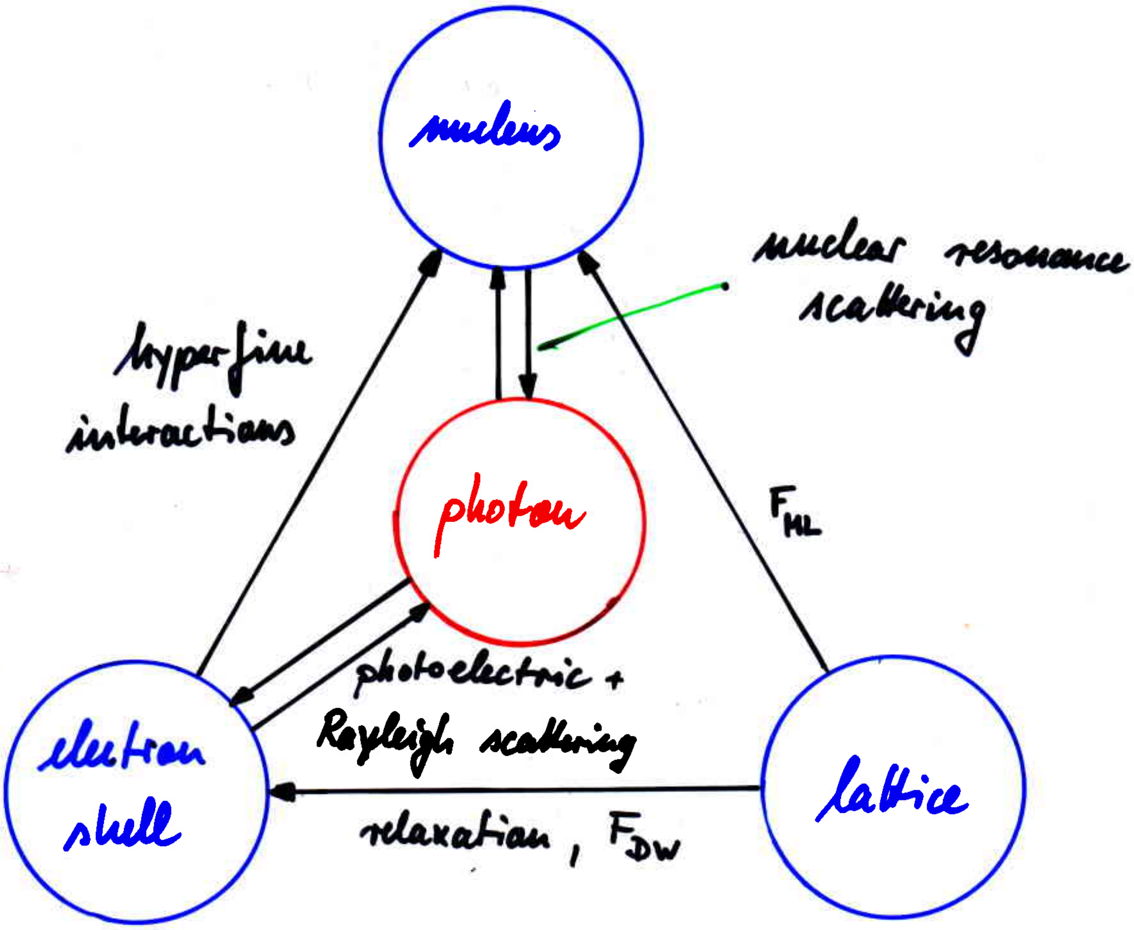
# NUCLEAR RESONANCE SCATTERING IN A FLUCTUATING ENVIRONMENT

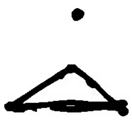
WOLFGANG STURHAHN

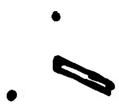
- Introduction
- Scattering Matrix
- Hyperfine Interactions
- Stochastical Model
- Numerical Treatment

# Scattering of X-rays in solids ...



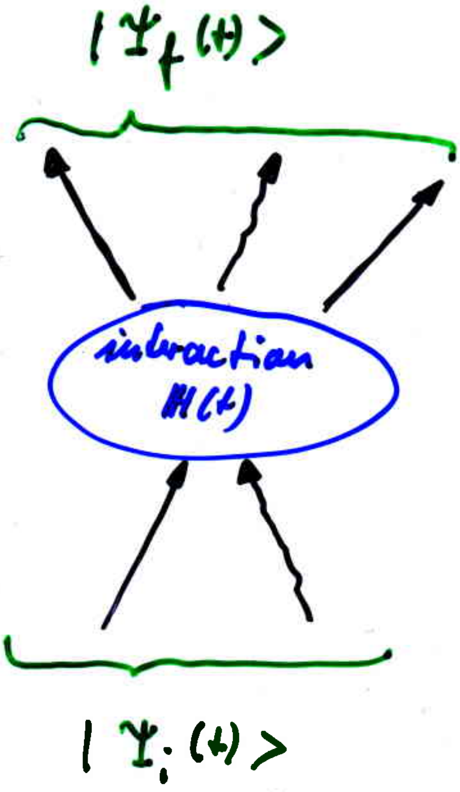

neglected:

 electronic thermal diffuse

 photon phonon

 nuclear inelastic

A scattering situation:




describe scattering by S-matrix:

$$S_{fi} = \lim_{t\to\infty} \langle \Psi_f(t) | \Psi_i(-t) \rangle$$

$$= \langle \Psi_f | S | \Psi_i \rangle$$

with

$$S = T\left\{ e^{-i\int dt\, \mathbb{H}(t)} \right\}$$

$$= \mathbb{1} - i\int dt\, \mathbb{H}(t) + \frac{(i)^2}{2!}\int dt\,dt'\; T(\mathbb{H}(t)\mathbb{H}(t')) + \ldots$$

The propagation of a photon is then described by

$$D^{fi}_{\mu\nu}(x,y) = -\frac{i}{\hbar c}\,\frac{\langle 0,\Psi_f|\, T\{S\, A_\mu(x)\, A_\nu(y)\}\,|\Psi_i, 0\rangle}{\langle 0,\Psi_f|\, S''\, |0,\Psi_i\rangle}$$

now introduce a matrix $M^{fi}_{\mu\nu}(x,y)$ which is determined by the properties of the scatterer

$$D^{fi}_{\mu\nu}(x,y) = D^{(0)}_{\mu\nu}(x,y) + \frac{i}{\hbar}\int dx'\,dy'\; D^{(0)}_{\mu\mu'}(x,x')\, M^{fi}_{\mu'\nu'}(x',y')\, D^{(0)}_{\nu'\nu}(y',y)$$

with

$$D^{(0)}_{\mu\nu}(x,y) = i\hbar c\, g_{\mu\nu}\cdot(-4\pi)\cdot\underbrace{\int \frac{e^{-ik(x-y)}}{k^2+i\varepsilon}\,\frac{d^4k}{(2\pi)^4}}_{=\delta_+(x-y)}$$

$$\rightsquigarrow\; D^{fi}_{\mu\nu}(x,y) = i\hbar c\, g_{\mu\nu}\,\delta_+(x-y) - i\hbar c^2\int dx'\,dy'\;\delta_+(x-x')\,\underbrace{M^{fi}_{\mu\nu}(x',y')\,\delta_+(y'-y)}_{\text{scattered contribution}}$$

in momentum representation

we get ...

$$\tilde{D}^{fi}_{\mu\nu}(k,-q) = i\hbar c\, g_{\mu\nu}(2\pi)^4\, \delta(k-q)\, \tilde{\delta}_+(k)$$

$$- i\hbar c^2\, \tilde{\delta}_+(k)\, \tilde{M}^{fi}_{\mu\nu}(k,-q)\, \tilde{\delta}_+(q)$$

and for the field of the scattered photon

$$A^{(S)}_\mu(k) = -\frac{c\,\tilde{\delta}_+(k)}{(2\pi)^4} \int dq\, \tilde{M}^{fi}_{\mu\nu}(k,-q)\, A^{(0)}_\nu(q)$$

incident field

with $\tilde{\delta}_+(k) = -\dfrac{4\pi}{k^2+i\varepsilon}$

in case of nuclear resonance scattering
the following scattering matrix is evaluated:

$$\tilde{M}^{fi}_{\mu\nu}(\vec{k},\omega;\vec{k}',\omega') = \frac{2\pi i}{c^3}\,\delta(\omega-\omega'+\omega_{fi})\,e^{i(\vec{k}'-\vec{k})\vec{R}} \times$$

$$\bar{K}_{fi}(\vec{k},\vec{k}') \times$$

$$\int_0^\infty dt\; e^{(i\omega-\Lambda)t} \langle\phi_f|\,\mathcal{J}_\mu(-\vec{k},t)\,\mathcal{J}_\nu(\vec{k}')\,|\phi_i\rangle$$

dependent on lattice vibrations:

$$\ldots = \langle\chi_f|\,e^{-i\vec{k}\vec{r}}\,|\chi_i\rangle\langle\chi_i|\,e^{i\vec{k}'\vec{r}}\,|\chi_i\rangle$$

fourier transforms in the cms of the nucleus

$$\mathcal{J}_\mu(-\vec{k},t) = e^{-i\vec{k}\vec{R}}\;e^{-i\vec{k}\vec{r}(t)} \times$$

$$\int d^3x\; e^{-i\vec{k}\vec{x}}\,j_\mu(\vec{x},t)$$

the time dependence of the nuclear currents may be expressed in terms of the hyperfine interaction operator $\mathbb{H}(t)$.

$$\mathcal{J}_\mu(-\vec{k},t) = \mathcal{U}^+(t)\,\mathcal{J}_\mu(-\vec{k})\,\mathcal{U}(t)$$

with

$$\mathcal{U}(t) = T\left\{ e^{-i\int_0^t \mathbb{H}(t')\,dt'} \right\}$$

the explicit time dependence of the hyperfine interaction operator is described by

$$\mathbb{H}(t) = -\frac{e^2}{c^2}\int \frac{j_\mu(\vec{x})\, S^\mu(\vec{x}',t)}{|\vec{x}-\vec{x}'|}\, d^3x\, d^3x'$$

$$\underbrace{+\,4\pi\,\frac{e^2}{c^2}\,Z\cdot\int\frac{S^0(\vec{x},t)}{|\vec{x}|}\,d^3x}_{\text{point charge interaction}}$$

after performing a multipole expansion we obtain

$$\mathbb{H}(t) = \sum_{L=0}^{2I'-1} \mathbb{H}^{(L)}(t)$$

with $\mathbb{H}^{(0)}(t) = \frac{2\pi}{3}\cdot\frac{e^2}{c^2}\cdot S^\mu_R(0,t)\int|\vec{x}|^2 j_\mu(\vec{x})\,d^3x$ ~ isomer shift

$$\mathbb{H}^{(1)}(t) = e\cdot\sum_M Q_{1M\mu}\,V^\mu_{1,-M}(t)\,(-1)^M$$ ~ magnetic dipole interact.

$$\mathbb{H}^{(2)}(t) = e\sum_M(-1)^M\,Q_{2M\mu}\,V^\mu_{2,-M}(t)$$ ~ electric quadrupole interact.

the time dependence of $\mathbb{H}$ and the time development operator $\mathcal{U}(t)$

$$\mathcal{U}(t) = T\left\{ e^{-i\int_0^t \mathbb{H}(t')\,dt'} \right\}$$

is of interest.

In the general case very complicated often even unknown time dependences have to be handled.

We consider three examples of simple time behaviour:

(1) fast relaxation

(2) slow relaxation

(3) fast switching

(1) fast relaxation case

$$\mathcal{U}(t+\Delta t) = T\left\{ e^{-i\int_t^{t+\Delta t} \mathbb{H}(t')\,dt'} \right\} \mathcal{U}(t)$$

if the time interval $\Delta t$ can be chosen that $\Delta t \cdot \Lambda \ll 1$ and $\mathbb{H}(t)$ is fluctuating sufficiently fast:

$$\int_t^{t+\Delta t} \mathbb{H}(t')\,dt' = \Delta t \cdot \bar{\mathbb{H}}$$

this implies

$$\int_0^{\varepsilon} \mathbb{H}(t'+\Delta t)\,dt' = \int_0^{\varepsilon} \mathbb{H}(t')\,dt' = \bar{\mathbb{H}}$$

for $\varepsilon \ll \Delta t \ll \frac{1}{\Lambda}$

$$\rightsquigarrow \quad \mathcal{U}^{+}(t)\, \mathbb{J}_{\mu}\, \mathcal{U}(t) = e^{i\bar{\mathbb{H}}t}\, \mathbb{J}_{\mu}\, e^{-i\bar{\mathbb{H}}t}$$

with $\bar{\mathbb{H}}|n\rangle = \Omega_n |n\rangle$ we get:

$$\mathcal{U}^{+}(t)\, \mathbb{J}_{\mu}\, \mathcal{U}(t) = \sum_{nn'} e^{i(\Omega_n - \Omega_{n'})t} |n\rangle\langle n| \mathbb{J}_{\mu} |n'\rangle\langle n'|$$

the energy dependence of scattering is then

$$\tilde{M}^{fi}_{\mu\nu}(\vec{k},\omega;\vec{k}',\omega') = \frac{2\pi}{c^3}\,\delta(\omega-\omega'+\omega_{fi})\; e^{i(\vec{k}'-\vec{k})\vec{R}} \times$$

$$\bar{K}_{fi}(\vec{k},\vec{k}') \times$$

$$\sum_n \frac{\langle\phi_f|\,J_\mu(-\vec{k})\,|\phi_n\rangle\langle\phi_n|\,J_\nu(\vec{k}')\,|\phi_i\rangle}{(\Omega_n+\varepsilon_f-\Omega_i-\varepsilon_i)-\omega'-i\Lambda}$$

which is the well known sum of lorentzians with widths of $2\Lambda$ and positions

$$(\Omega_n+\varepsilon_f-\Omega_i-\varepsilon_i)$$

lattice energies

(2) slow relaxation case

$$\mathbb{U}(t) = e^{-i\,\mathbb{H}(0)\,t}$$

here the Hamiltonian does not change within a few nuclear lifetimes.

But $\mathbb{H}(0)$ takes different values for different nuclei.

This looks as if a certain amount of sublattices exist each with its own specific hyperfine interaction.

(3) fast switching

$$\mathbb{H}(t) = \begin{cases} \mathbb{H}_< & \text{for} \quad t \le \tau \\ \mathbb{H}_> & \text{for} \quad t > \tau \end{cases}$$

$\tau$ ~ switching time

$$\rightsquigarrow \quad \mathcal{U}(t) = \begin{cases} e^{-i\mathbb{H}_< t} & \text{f.} \quad t \le \tau \\ e^{-i\mathbb{H}_< \tau}\, e^{-i\mathbb{H}_> (t-\tau)} & \text{f.} \quad t > \tau \end{cases}$$

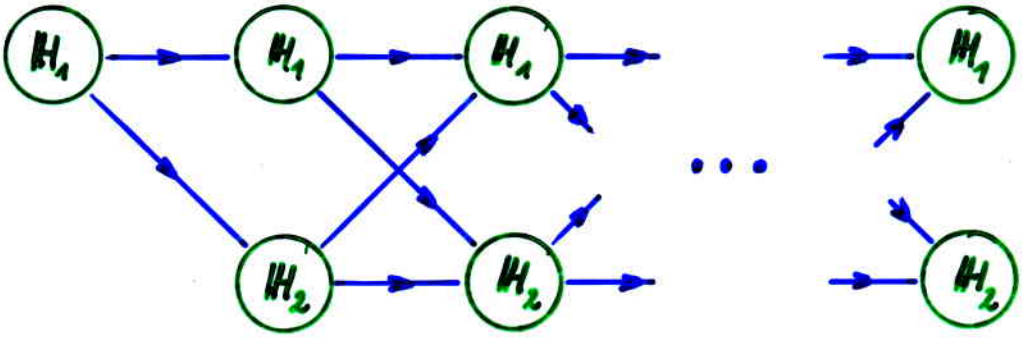
the time dependent nuclear currents then evaluate to be

$$\mathcal{J}_\mu(t) = \begin{cases} e^{i\mathbb{H}_< t}\, \mathcal{J}_\mu(0)\, e^{-i\mathbb{H}_< t} & \text{f.} \quad t \le \tau \\ e^{i\mathbb{H}_> (t-\tau)}\, \mathcal{J}_\mu(\tau)\, e^{-i\mathbb{H}_> (t-\tau)} & \text{f.} \quad t > \tau \end{cases}$$

the calculation of the energy dependence of the scattering in this case is straightforward...

The hyperfine interaction is described by

$\mathbb{H}_1$ or $\mathbb{H}_2$ dependent on time




$t_0 = 0$ $\quad t_1 = \Delta t$ $\quad t_2 = 2\Delta t$ $\quad \dots$ $\quad t_n = n \cdot \Delta t$

paths : $\{\mathbb{H}\}^n$

probability for path j : $P(\{\mathbb{H}\}_j^n)$ ; $\sum_{\{j\}^n} P(\{\mathbb{H}\}_j^n) = 1$

path integral : $\mathbb{I}_j^n = \sum_{l=0}^{n} \mathbb{H}_j(t_l) \cdot \Delta t$

stochastical average :

$$\langle F(t) \rangle_{av} = \lim_{\substack{n \to \infty \\ (t = n \cdot \Delta t)}} \sum_{\{j\}^n} P(\{\mathbb{H}\}_j^n) \cdot F(\mathbb{I}_j^n)$$

$$= \sum_{\alpha\beta} \underbrace{\lim_{n \to \infty} \sum_{\{j\}^{n-2}} P(\{\mathbb{H}\}_{\alpha j \beta}^n) \, F(\mathbb{I}_{\alpha j \beta}^n)}_{G_{\alpha\beta}(t)}$$

consider a stationary Markoff process ...

$$P(\{H\}_1^n) = P(H_{j_0}(t_0), t_0) \cdot W(H_{j_0}(t_0) \mid H_{j_1}(t_1), \Delta t) \ldots$$
$$W(H_{j_{n-1}}(t_{n-1}) \mid H_{j_n}(t_n), \Delta t)$$

introduce :

$$W_{\beta\gamma}(\Delta t) = W(H_\beta \mid H_\gamma, \Delta t) = \delta_{\beta\gamma} + \lambda_{\beta\gamma} \cdot \Delta t$$

with $\sum_\gamma W_{\beta\gamma} = 1$, $\sum_\gamma \lambda_{\beta\gamma} = 0$

transition probabilities

and ...

$$F(\mathbb{I}^{n+1}_{\alpha j \beta\gamma}) = F(\mathbb{I}^n_{\alpha j \beta} + H_\gamma \Delta t)$$

$$= F(\mathbb{I}^n_{\alpha j \beta}) + D^n_{\alpha j \beta\gamma} \cdot \Delta t$$

derivation of $F(t)$

„equation of motion" :

$$\frac{d}{dt} G_{\alpha\beta}(t) = \sum_{\gamma} G_{\alpha\gamma}(t)\, \lambda_{\gamma\beta} + \sum_{\{j\}^{n-2}} P(\{\mathbb{H}\}^{n}_{\alpha j \beta})\, D^{n}_{\alpha j \beta\beta}(t)$$

in our case we have

$$D^{n}_{\alpha j \beta\beta}(t) = i\, \mathbb{H}_{\beta}\, F(\mathbb{I}^{n}_{\alpha j \beta}) - i\, F(\mathbb{I}^{n}_{\alpha j \beta})\, \mathbb{H}_{\beta}$$

and we get

$$\frac{d}{dt} G_{\alpha\beta}(t) = \sum_{\gamma} G_{\alpha\gamma}(t)\, \lambda_{\gamma\beta} + i\, \mathbb{H}_{\beta}\, G_{\alpha\beta}(t) - i\, G_{\alpha\beta}(t)\, \mathbb{H}_{\beta}$$

with the starting condition

$$G_{\alpha\beta}(0) = p_{\alpha}\, \delta_{\alpha\beta}\, \mathbb{J}_{\mu}(-\vec{k})$$

solution of the „equation of motion" :

introduce matrix elements

$$\langle I m | H_\beta | I' m' \rangle = \delta_{II'} \, H_\beta^{I m m'}$$

$$\langle I m | G_{\alpha\beta} | I' m' \rangle = G_{\alpha\beta}^{II' m m'}$$

$$\langle I m | \vec{j}_\mu(-\vec{k}) | I' m' \rangle = \sqrt{\frac{4\pi c \Lambda}{k}} \times$$

$$\sum_{L\lambda} \Delta_{L\lambda} \cdot C(I L I'; m\; m'-m) \cdot \left[ \vec{Y}^{(\lambda)}_{L, m'-m}(\hat{k}) \right]_\mu$$

for a pure $(L\lambda)$ multipole transition
( e.g. $M1 \mathrel{\hat{=}} L=1, \lambda=0$ for $^{57}Fe$, $^{169}Tm$, $^{119}Sn$ )
the last term simplifies to

$$\langle I m | \vec{j}_\mu | I' m' \rangle = \sqrt{\frac{4\pi c \Lambda}{k}} \cdot \Delta_{L\lambda} \times$$

$$C(I L I'; m\; m'-m) \cdot \left[ \vec{Y}^{(\lambda)}_{L, m'-m}(\hat{k}) \right]_\mu$$

$$\leadsto \quad \dot{G}_{\alpha\beta}^{II' m m'}(t) = -i \sum_{\gamma M M'} A_{\gamma\beta}^{II' m m' M M'} \; G_{\alpha\gamma}^{II' M M'}(t)$$

with $$A_{\gamma\beta}^{II' m m' M M'} = i\, \lambda_{\gamma\beta}\, \delta_{mM}\, \delta_{m'M'} - H_\beta^{I m M}\, \delta_{\beta\gamma}\, \delta_{m'M'}$$

$$+ H_\beta^{I' M' m'}\, \delta_{\beta\gamma}\, \delta_{mM}$$

this differential equation solves immediately

$$G_{\alpha\beta}^{II'mm'}(t) = \sum_{\gamma MM'} \left( e^{-i\underline{\underline{A}}^{II'} \cdot t} \right)_{\gamma\beta}^{mm'MM'} \cdot G_{\alpha\gamma}^{II'MM'}(0)$$

$$= \rho_\alpha \cdot \sqrt{\frac{4\pi c \Lambda}{k}} \, \Delta_{L\lambda} \times$$

$$\sum_{MM'} \left( e^{-i\underline{\underline{A}}^{II'} \cdot t} \right)_{\alpha\beta}^{mm'MM'} \cdot C(ILI'; M\, M'-M) \cdot \left[ \vec{Y}_{L,M'-M}^{(\lambda)} \right]_\mu$$

and we get for the stochastical average

$$\left\langle \langle Im | \mathcal{U}^+(t)\, \mathcal{T}_\mu\, \mathcal{U}(t) | I'm' \rangle \right\rangle_{av} = \sum_{\alpha\beta} G_{\alpha\beta}^{II'mm'}(t)$$

$$= \sqrt{\frac{4\pi c \Lambda}{k}} \cdot \Delta_{L\lambda} \times$$

$$\sum_{\alpha\beta MM'} \rho_\alpha \left( e^{-i\underline{\underline{A}}^{II'} \cdot t} \right)_{\alpha\beta}^{mm'MM'} C(ILI'; M\, M'-M) \times$$

$$\left[ \vec{Y}_{L,M'-M}^{(\lambda)} \right]_\mu$$

the numerical treatment focusses on
the solution of the eigenwert problem :

$$\sum_{\substack{jj'll' \\ \gamma\varepsilon}} L^{mm'jj'}_{\alpha\gamma} \; A^{jj'll'}_{\gamma\varepsilon} \; R^{ll'\mu\mu'}_{\varepsilon\beta} = \Omega^{mm'}_{\alpha} \cdot \delta_{\alpha\beta} \, \delta_{m\mu} \, \delta_{m'\mu'}$$

left eigenvectors — right eigenvectors — eigenvalues

the dimension of the matrices is

$$\underbrace{(2I+1)}_{\text{spin of nuclear ground state}} \times \underbrace{(2I'+1)}_{\text{spin of nuclear excited state}} \times \underbrace{N}_{\text{number of external states}}$$

e.g. : $^{57}Fe_{5/2}$

$$2I+1 = 2$$
$$2I'+1 = 4$$
$$N = 6$$

$$\leadsto (2I+1)(2I'+1)\,N = 48$$

After the EW problem has been solved numerically we write

$$A_{\alpha\beta}^{mm'MM'} = \sum_{jj'\gamma} R_{\alpha\gamma}^{mm'jj'} \Omega_\gamma^{jj'} L_{\gamma\beta}^{jj'MM'}$$

and

$$\left(e^{-i\underline{\underline{A}}^{II}\cdot t}\right)_{\alpha\beta}^{mm'MM'} = \sum_{jj'\gamma} R_{\alpha\gamma}^{mm'jj'} e^{-i\Omega_\gamma^{jj'}t} L_{\gamma\beta}^{jj'MM'}$$

note: $\Omega_\gamma^{jj'}$ values are not real since $\underline{\underline{A}}$ is not hermitian

in detail ..

$$A_{\alpha\beta}^{mm'MM'} - \left(A_{\beta\alpha}^{m'mM'M}\right)^* = i\left(\lambda_{\alpha\beta} + \lambda_{\beta\alpha}\right)\delta_{mM}\delta_{m'M'}$$

this reflects the speed up of nuclear decay due to relaxation.

the nuclear scattering matrix is then given by

$$\tilde{M}^{(elastic)}_{\mu\nu} = \frac{k}{2}\,\sigma_0\,F_{ML}\cdot\sum_{\gamma jj'} \frac{\left[\vec{\mathcal{L}}^{(\lambda)\,jj'}_{L\gamma}(\hat{k})\right]_\mu \left[\vec{\mathcal{R}}^{(\lambda)\,jj'}_{L\gamma}(\hat{k}')\right]_\nu}{z^{jj'}_\gamma(\omega) - i}$$

with $$z^{jj'}_\gamma(\omega) = \frac{1}{\Lambda}\left(\Omega^{jj'}_\gamma - \omega\right)$$

$$\vec{\mathcal{L}}^{(\lambda)\,jj'}_{L\gamma}(\hat{k}) = \sqrt{\frac{8\pi}{2I'+1}}\sum_{\beta MM'} L^{jj'MM'}_{\gamma\beta}\,C(ILI';M\,M'-M)\,\vec{Y}^{(\lambda)}_{L,M'-M}(\hat{k})$$

$$\vec{\mathcal{R}}^{(\lambda)\,jj'}_{L\gamma}(\hat{k}') = \sqrt{\frac{8\pi}{2I'+1}}\cdot\sum_{\beta MM'} p_\beta\,R^{MM'jj'}_{\beta\gamma}\,C(ILI';M\,M'-M)\,\vec{Y}^{(\lambda)\,*}_{L,M'-M}(\hat{k}')$$

the effects of multiple scattering are obtained by the usual procedure starting at the scattering matrix of the MB atom.

- the rules of QED have been applied to calculate nuclear resonance scattering

- time dependent hyperfine interactions were considered by use of a stochastical model

- an expression similar to that used for stationary hyperfine interactions was derived