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SOME NEW RESULTS IN MÖSSBAUER RELAXATION THEORY

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Abstract.- We discuss the validity of the perturbation treatments and we derive some expressions for the Mössbauer lineshape both without and with the noise approximation.

VALIDITY RANGE AND COMPARISON OF VARIOUS TREATMENTS

The problem of relaxation effects in Mössbauer spectra has given rise to several approaches based on a) stochastic models b) the relaxation equation for the density matrix c) the resolvent method following Fano. All three approaches use Liouville formalism and lead to relaxation supermatrices which enter the Mössbauer lineshape. Methods b) and c) have been compared in great detail in Ref. [11] : we summarize here the conclusions of this study.

Both b) and c) are perturbation methods. It can be shown that if the "white noise approximation" (WNA) is valid they can describe the whole range of relaxation effects in the Mössbauer spectra (small, intermediate or large compared with the distance between lines). On the contrary if the WNA is not valid, they are only valid for small relaxation effects and then one can apply the "secular approximation". This last result solves the ambiguities met in p. 301 of [2] (note), in p. 307 of [2] (top of first column) and in p. 2117 of [2] (the spurious third order terms always cancel out when the theory applies).

Method b) leads to relaxation supermatrices $R$ and $S$, while c) leads to $R'(w+i\eta)$ and $S'(w+i\eta)$.

When the WNA is valid $R=R'$, $S=S'$. When the WNA is not valid it can be shown that the matrix elements of $R$ and $R'$ (or $S$ and $S'$) which control the broadening of a given Mössbauer line become identical at the top of that line. I.e., line profiles obtained with both methods are very similar, although c) is slightly better.

EXPRESSIONS FOR THE MOSSBAUER LINESHAPE WHEN THE WNA IS NOT VALID

I/ Lineshape in the isotropic case. In Ref. [1] we used the tensor operator method in order to compute the Mössbauer lineshape for an ion with a hyperfine coupling $A_1 \cdot \vec{S}$ in the excited state and $A_2 \cdot \vec{S}$ in the ground state, when its electronic spin $\vec{S}$ is submitted to a coupling $-g_\mu B H\cdot \vec{S}$ with a random field $\vec{H}$ whose fluctuations are isotropic and are characterized by a correlation function:

$$H(t) = \langle H(t)H(0) \rangle$$

(this is a high temperature calculation : $k_B T >> A, A_g$).

Here we will not give the general formulas obtained in [1] but only their application to the Mössbauer lineshape of Yb$^{170}$ in the electronic doublet state $I_e=2$, whence two hyperfine states $I_g=5/2$ with $E_F=A$ and $F=3/2$ with $E_F=-3/2 A$, and a ground state $I_g=0$ with no h.f.s. Let us define:

- $X = I(w+3/2 \eta^2 + 1 I_g/2)$
- $Y = I(w-A/M + 1 I_g/2)$

( where $I(w)$ is defined above : Eq (1)). The Mössbauer lineshape of Yb$^{170}$ is found to be $(\rho=r/2-iw)$.
A formula equivalent to this one has been derived by Afanasev et al. 131. When the WNA is valid, Eq(2) reduces to Eq(34) of 4 with X + Y = 1/2 T1S.

2/ Linesshape of (Yb170)3+ in uniaxial symmetry. Assume that state I = 2 has a hyperfine structure: 

\[ \mathcal{H}_0 = A / L S_z + a(I^2 - 2) \]  

with \( S = 1/2 \) and that \( S \) is submitted to a fluctuating field \( \mathcal{H} \) such that \( S = \sum \mathcal{H} \mathcal{H}_z(t) \mathcal{S}_z \). \( m \) being the eigenvalue of \( L \).

The PAC result for the damping coefficient of the multipole of order \( L \) (Eq. (VI) 121 with \( k = L \).

We want to compute \( \gamma \). As a check, when \( I = 3/2, L = 1 \) we recover the Bradford Marshall result. For b/R we recover the Bradford Marshall result [6] for b/R of the excited state I = 3/2.

\[ \gamma = \frac{3}{2} \mathcal{H}_0 \mathcal{H}_z(t) \mathcal{S}_z(T) \]  

When the WNA is valid \( \mathcal{W}_0 = \mathcal{W}_L \), and this expression reduces to Eq(79) of \(|5|\).

WNA VALID : ANALOG FOR THE M.E. OF ABRAGAM-POUND COEFFICIENTS FOR PERTURBED ANGULAR CORRELATIONS

When relaxation is very fast the spectrum reduces to a single broadened line \( \mathcal{J}(\omega) = \text{Re} \left[ \frac{1}{1 - i \omega / T + 2 \delta} \right] \).

We want to compute \( \delta \).

1/ Paramagnetic case. Assume a Mössbauer transition with multipolarity \( L \) and a fluctuating Hamiltonian \( A \mathcal{I}_z \mathcal{S}_z(t) \) or \( A \mathcal{I}_z \mathcal{S}_z(t) \) with correlation time \( T_1 S \) (electronic relaxation time). Tensor operator methods lead to:

\[ \delta = \frac{S(T + 1)}{3h^2} T_1 S \left( \text{A}^2 \mathcal{I} \mathcal{I} + \text{A}^2 \mathcal{I} (I^2 + 1) \right) \]  

where \( \text{A}^2 \mathcal{I} \mathcal{I} = (\mathcal{I} - \frac{1}{2})^2 \mathcal{I} \mathcal{I} \) \( \mathcal{I} \) being the eigenvalue of \( \mathcal{I} \).

As a check, when \( I = 3/2, I = 1/2 \) and \( L = 1 \) \( \delta = \frac{3}{2} \mathcal{H}_0 \mathcal{H}_z(T) \mathcal{S}_z(T) \)


\[ \delta = \frac{S(T + 1)}{3h^2} T_1 S \left( \text{A}^2 \mathcal{I} \mathcal{I} + \text{A}^2 \mathcal{I} (I^2 + 1) - \frac{1}{2} \mathcal{I} \mathcal{I} \right) \]  

On the other hand, when \( A = \text{A}, I = I \), we recover the PAC result for the damping coefficient of the multipole of order \( L \) (Eq. (VII) 42) of \( \delta = \frac{3}{2} \mathcal{H}_0 \mathcal{H}_z(T) \mathcal{S}_z(T) \) with \( k = L \).