Oxidation in ^{57}Fe Compounds Using Mossbauer Spectroscopy

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Two big problems with γ-ray spectroscopy:

1. Doppler broadening from thermal motion of atoms
2. Recoil of emitting atom displaces emission line from absorption line

Mossbauer emission: Line broadening and recoil characterized by emission in crystal lattice.

Outline

- Recoil energy R from a γ-ray given by

$$R = E_\gamma^2/2mc^2 \tag{1}$$

- Momentum transfer but not energy transfer $\rightarrow$ confinement to lattice.

- Fraction undergoing recoilless emission:

$$\begin{aligned} P\left(N_i \rightarrow N_f, L_i \rightarrow L_f\right) &\propto \left| \langle f| H_{\text{int}} |i\rangle \right|^2 \\ &= \left| \langle L_f| e^{i\mathbf{k}\cdot\mathbf{r}} |L_i\rangle \langle N\rangle \right|^2, \end{aligned} \tag{2}$$

For a harmonic oscillator model ($R \ll \hbar\omega$):

$$f = \left| \langle L_f| e^{i\mathbf{k}\cdot\mathbf{r}} |L_i\rangle \right|^2 = e^{-R/\hbar\omega_E} \tag{3}$$

- Probability of recoilless emission from a solid lattice:

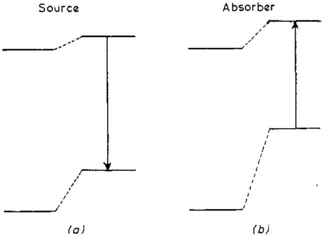
$$P\left(E\right) = \frac{\Gamma/2\pi}{\left(E - E_0\right)^2 + \frac{1}{4}\Gamma^2} \tag{4}$$

- With moving source, γ-ray doppler shifted:

$$\frac{\Delta E}{E} = \frac{v}{c} = \beta, \tag{5}$$

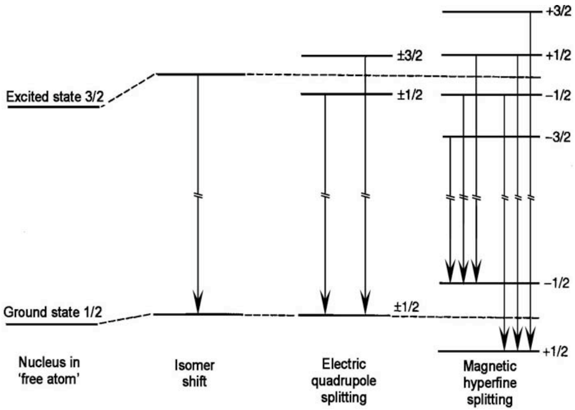
allowing us to sweep through resonance absorptions.

- Energy level shift between source and absorber:

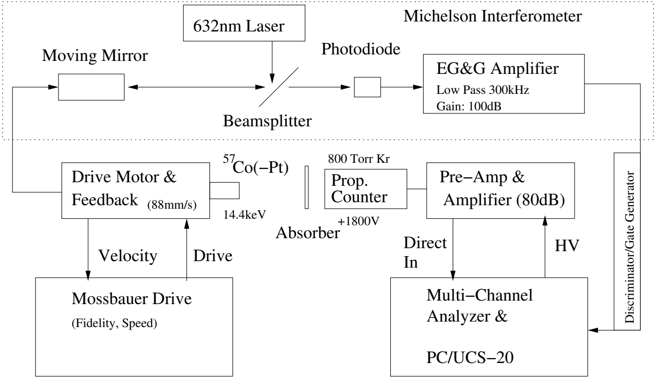


- Velocity to restore resonance,

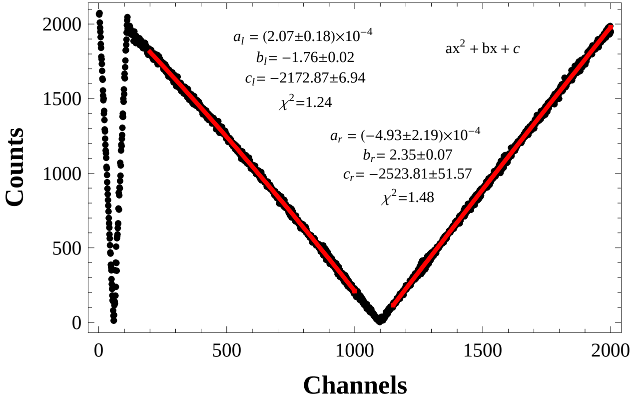
$$\delta = \Delta E_A - \Delta E_S \propto \left(\langle R_e^2 \rangle - \langle R_g^2 \rangle \right) \left(|\psi_s(0)|_A^2 - |\psi_s(0)|_S^2 \right) \tag{6}$$



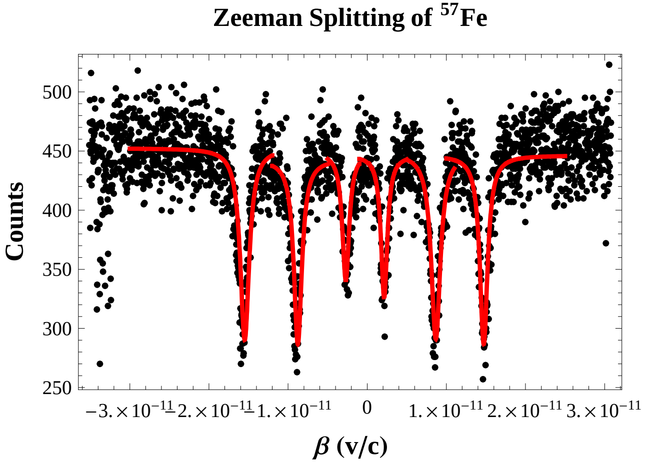
- Using $E_n = -g_n \mu_N IH + \frac{1}{2}\epsilon + \delta_n$: Zeeman/quadrapole splitting, polarization, isomer shift, and magnetic moments.
- Peak height ratios 3 : β : 1 : 1 : β : 3: $\beta = 4\sin^2\theta / \left(1 + \cos^2\theta\right)$



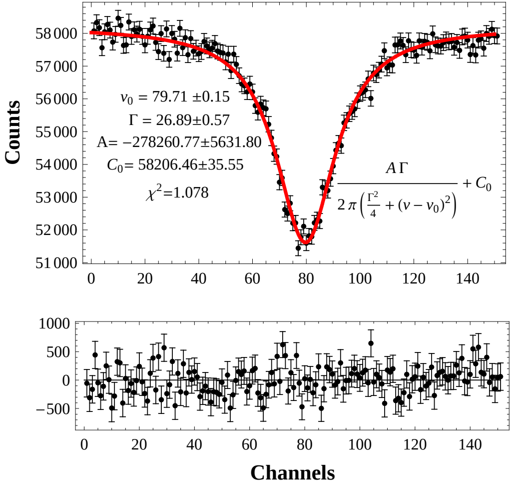
Parameters: Temperature, absorber composition and thickness, source distance.



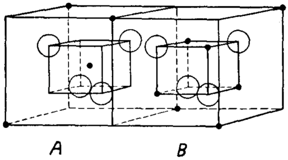
Velocity and counts related by $v_i = \frac{C_i \lambda}{2NT}$


Zeeman Splitting of ^{57}Fe
Counts
500
450
400
350
300
250
$-3.\times10^{-11}$
$-2.\times10^{-11}$
$-1.\times10^{-11}$
0
$1.\times10^{-11}$
$2.\times10^{-11}$
$3.\times10^{-11}$
β (v/c)

Sample Fit and Residuals for Fe_2O_3



- $Fe_2^{+3}O_3$ and $Fe_2^{+3}Fe^{+2}O_4$ (tetrahedral and octahedral)
- Fe_3O_4: composite spectrum of two oxidation states.



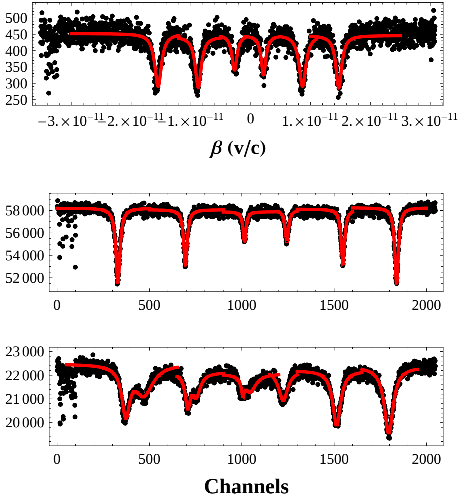


Table: Properties of iron Mossbauer absorbers.

	Fe_2O_3		Fe_3O_4 (tetrahedral)		Fe_3O_4 (octahedral)	
	Measured	Published	Measured	Published	Measured	Published
g_0'[mm/s]	6.17 ± 0.02	6.11 ± 0.05	6.26 ± 0.08	5.90 ± 0.2	5.81 ± 0.09	5.3 ± 0.2
g_1'[mm/s]	3.37 ± 0.04	3.45 ± 0.03	3.20 ± 0.10	3.35 ± 0.15	3.02 ± 0.07	3.1 ± 0.1
μ_1/μ_0[mm/s]	1.64 ± 0.02	1.69 ± 0.02	1.53 ± 0.04	1.7 ± 0.1	1.56 ± 0.03	1.75 ± 0.09
δ[mm/s]	0.38 ± 0.05	0.47 ± 0.03	0.36 ± 0.09	0.45 ± 0.1	0.58 ± 0.10	0.7 ± 0.1
ϵ[mm/s]	0.124 ± 0.003	0.12 ± 0.03	0.16 ± 0.05	0.0 ± 0.1	0.03 ± 0.01	0.0 ± 0.1
$H\,[T]$	53.5 ± 0.6	51.5	52.4 ± 0.5	50 ± 2	47.0 ± 0.6	45 ± 2
$\theta\,[^\circ]$	—	—	—	—	—	—

- Values determined through least-squares method.
- Polarization angle averages: $3 : \beta : 1$, where $\beta = \frac{4\sin^2\theta}{1+\cos^2\theta}$.

- Statistical counting error
- Michelson interferometer velocity calibration
- Finite velocity resolution of apparatus
- Finite source and absorber thickness
    - Gaussian line broadening
    - Saturation of line intensity
- Oxidation sample contamination

Summary

- Magnetism vs. Temperature: Curie temperature for ferromagnetic iron compounds?
- Sulfate samples missing/damaged (Replacements?).
- Sample impurity: Aside from magnetite Fe_3O_4, all are with 1-2σ of accepted values.