Mossbauer Spectroscopy for the Determination of Quadrupole Splitting and Isomer Shift

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Outline

1. Introduction, Motivations

2. Theory: Nuclear Resonance, Recoilless Emission, Mossbauer Spectroscopy, Quadrupole Splitting, Isomer Shift

3. Apparatus and Experiment

4. Results & Error Analysis: Calibrations, Comparisons with Known Data

5. Conclusions

Introduction: Why Mossbauer Spectroscopy?

- Interested in observing resonance absorption of nuclear radiation, as previously observed optically
- Recoil of nucleus gives a shift in energy too large to observe resonance absorption
- Mossbauer spectroscopy: recoilless emission allows us to see resonance absorption (1957)
- Can then probe properties of resonance, other effects

Nuclear Absorption and Emission

- For conservation of momentum, nucleus must recoil, has kinetic recoil energy

- Energy of the gamma ray absorbed or emitted has an energy difference of $\frac{E^2}{2mc^2}$, difference between absorbed and emitted is twice this, which is much larger than the linewidth, E is energy of the transition, m is nuclear mass, c is speed of light

- Don't see resonance

Recoilless Emission

- Instead, use a crystal lattice as a substrate
- Only recoil would come from phonons (vibrations of lattice), with quantized energies
- For recoil energy less than lowest phonon energy, $k\Theta_D$, no recoil, k Boltzmann constant, Θ_D Debye temperature
- See resonance absorption

Mossbauer Spectroscopy

- Moving source or emitter creates a Doppler shift $\frac{\Delta E}{E} = \frac{v}{c}$, E energy, v velocity, c speed of light

- By probing a range of velocities, we can shift in and out of the resonance energy

Quadrupole Splitting

- Quadrupole moment comes from a non-spherical nuclear charge distribution

- Quadrupole splitting arises from an internal electric field gradient

- $\Delta E = e\frac{3m_I^2 - I(I+1)}{4I(2I-1)}Q\frac{\partial^2 V}{\partial z^2}$, e is charge of electron, I is angular momentum, m_I magnetic quantum number of nuclear state, Q quadrupole moment, V electric potential

- For Fe-57, ground state, I=1/2, $\Delta E = 0$, excited state, I=3/2, $\Delta E = \pm\frac{1}{8}Qe\frac{\partial^2 V}{\partial z^2}$

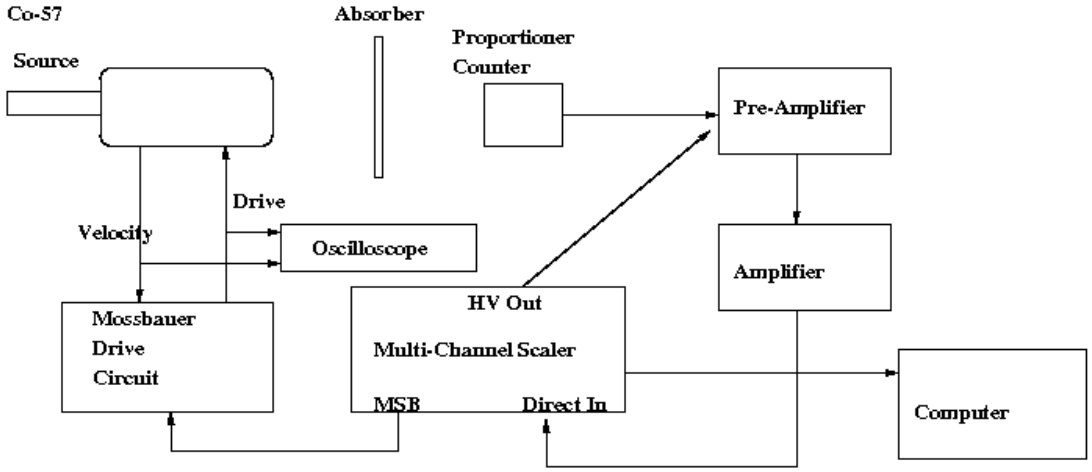
Isomer Shift

- Arises from changes in chemical environment: s electron wavefunctions overlap with nucleus

- Because nuclear charge is not a point charge, but a distribution, Coulomb potential is altered for different nuclear radii, affects nuclear energy levels

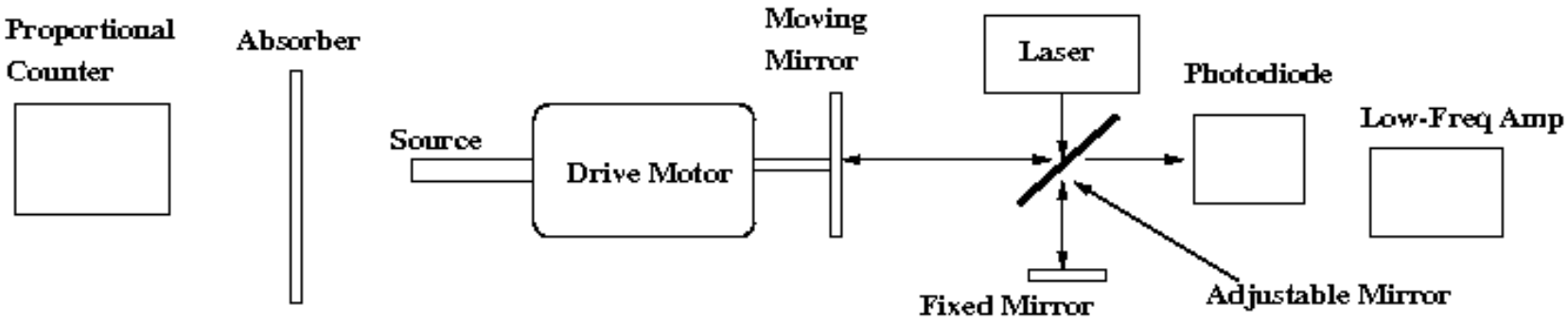
- $\Delta E = \frac{2\pi}{5} Z e^2 (R_{is}^2 - R_{gs}^2)(|\psi(0)_a|^2 - |\psi(0)_e|^2)$, Z nuclear charge, R radius, is excited state, gs ground state, $\psi(0)_a$ electron wavefunction of absorber, $\psi(0)_e$ electron wavefunction of emitter at R=0

Apparatus



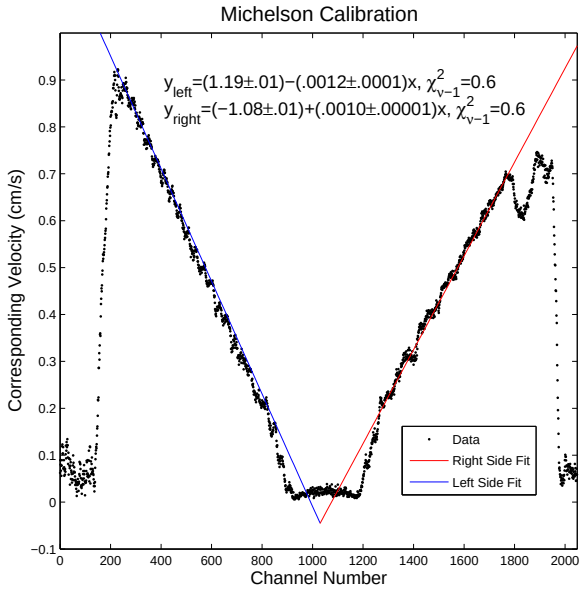
- Co-57 decays into excited Fe-57, emits without recoil, 14.4 keV photon
- Recoilless absorption causes a drop in the number of detector counts
- Sweep over a range of velocities to move in and out of resonance
- Observe a number of nuclear effects using different absorbers

Calibration

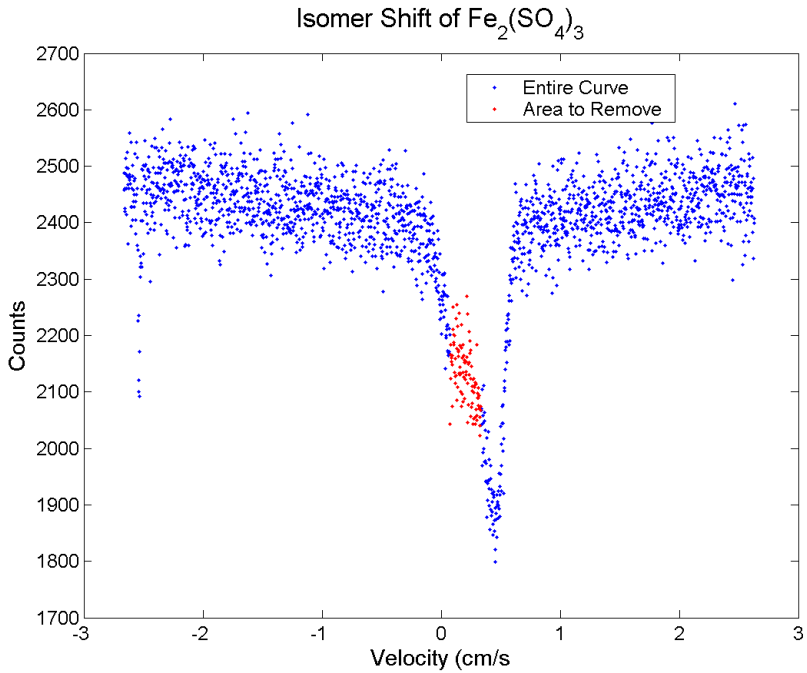


- Need to match channel number to a velocity
- Constructive interference when piston moves distance $\lambda/2$
- In any given channel, $V_i = \frac{C\lambda}{2NT}$, V is velocity, i is channel number, N is number of sweeps, T is dwell time, C is counts

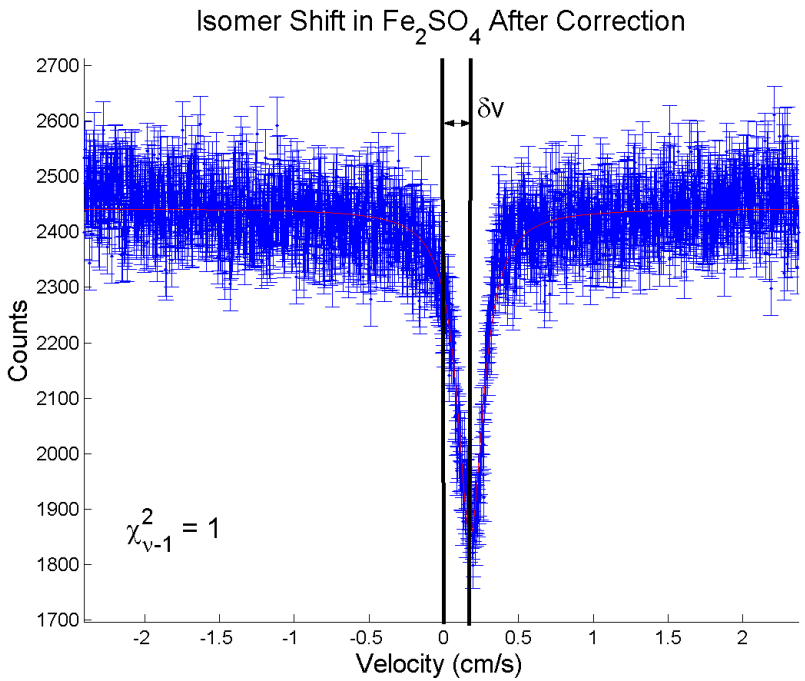
Calibration



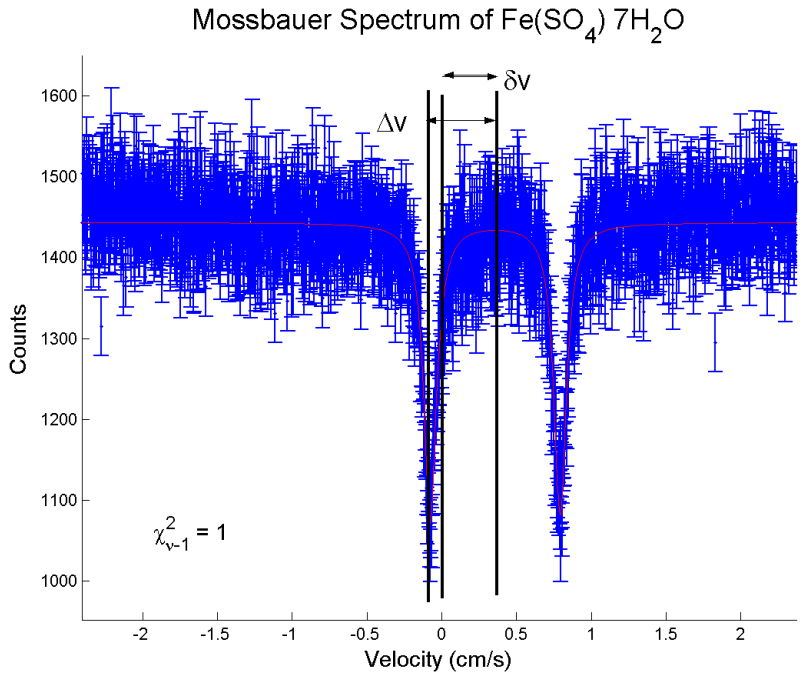
- Fit positive and negative velocities to two lines, don't intersect
- Calibration Equation: v=(x-a)b, v = velocity, x = channel number, a = (1030 ± 40), b = (0.0011 ± 0.0001)
- Use another spectrum as a secondary calibration (Zeeman)



- Resonant peak is not Lorentzian-reflective of bad drive velocity
- To correct, remove area where counts don't change: 100 ± 30 channels
- No quadrupole splitting because no field gradient for Fe^{+++} (it's a $3d^5$ state)



- Fit Lorentzian to resonant peak: perfect $\chi^2_{\nu-1}$
- Calculate velocity to be (0.18 $\pm$ 0.13) cm/s, δE = (8 $\pm$ 6) x 10^{-8} eV
- Very large error comes 99% from uncertainty in calibration coefficient because the peak is near zero velocity



- Fit 2 Lorentzians with same correction: perfect $\chi^2_{\nu-1}$
- Calculate quadrupole splitting from Fe^{++} ($3d^6$ state) of $\Delta v = (.4 \pm 0.1)$ cm/s, $\Delta E = (2.1 \pm 0.5)$ x 10^{-7} eV
- Isomer shift $\delta v = (0.3 \pm 0.1)$ cm/s, $\delta E = (1.7 \pm 0.5)$ x 10^{-7} eV

Error Analysis

- From Michaelson, v = (x-a)*b*c, v is velocity, a is zero of velocity in channels, x is channel number, c is scaling factor
- $\sigma_v^2 = v^2(\frac{\sigma_x^2+\sigma_a^2}{(x-a)^2} + \frac{\sigma_b^2}{b^2} + \frac{\sigma_c^2}{c^2})$
- For isomer shifts, a contributes 99% of σ
- For quadrupole shift, a contributes 61% to σ, b and c contribute about 19% each

Results and Interpretation

- Isomer shift $\delta E = (8 \pm 6)$ x 10^{-8} eV for $Fe_2(SO_4)_3$
- From literature, should be 2.4 x 10^{-8} eV
- Isomer shift $\delta E = (1.7 \pm 0.5)$ x 10^{-7} eV for $Fe(SO_4)$ $7H_2O$
- From literature, expect 6.7 x 10^{-8} eV
- Quadrupole splitting $\Delta E = (2.1 \pm 0.5)$ x 10^{-7} eV for $Fe(SO_4)$ $7H_2O$
- From literature, should be 1.5 x 10^{-7} eV

Conclusions

- Observe isomer shift and quadrupole splitting of iron-containing samples

- For better results, need to improve treatment of velocities around zero