THE MÖSSBAUER EFFECT

Resonant gamma ray fluorescence is a useful tool in determining a variety of nuclear and solid state properties. The discovery of the Mössbauer effect greatly increased the accuracy and capabilities of gamma ray fluorescence. This in turn has made possible the accurate measurement of nuclear lifetimes, nuclear magnetic moments, and nuclear spins. In this lab you will use resonant gamma ray fluorescence and the Mössbauer effect to determine the magnetic moment of the first excited state of Fe⁵⁷ and measure the bulk internal magnetic field in natural iron.

Introduction: Energy Scales

There are several energies that characterize gamma ray fluorescence. The first is the *excitation energy*, E_0 , of the nuclear transition being studied. This is usually the energy difference between a nucleus in an excited state and in its ground state. When a nucleus decays from an excited state, the excitation energy is often radiated away as a gamma ray. In practice, the energies of such gamma rays are typically less than 1 MeV. [1]

Due to quantum mechanics, there is a small spread in the possible energy values of the emitted gamma ray. The uncertainty principle states that

$$\Delta E_0 \Delta t \ge \hbar \tag{1}$$

where \hbar is Planck's constant and Δt is the time taken to measure the energy level. It is important to note that Δt is NOT the same as the lifetime of the state, τ . Assuming one can measure the the excited state for no more than the lifetime τ we get an lower bound on the energy spread ΔE_0 . This energy spread is called the *linewidth* and is denoted Γ . Experimentally it is determined by measuring the width of an energy peak at its half maximum. See Fig. 1 a,b. The energy scale for Γ is usually many orders of magnitude smaller than E_0 . [1]

INSERT FIGURE HERE

Figure 1. a) Energy of transitions for a nucleus emitting a gamma ray. The vertical scale indicates the probability of a transition. Notice that E_0 is the only possible transistion. b) the natural linewidth makes a small spread in the possible energies of transition. c) energy splitting shifts the single peak into two slightly separated peaks. d) energy loss due to recoil causes both peaks to shift to a slightly lower energy level. In the case of absorbtion, nuclear recoil requires an upward shift in the energy of the exciting gamma ray.

The next energy scale of interest is determined by the experiment being done. An example is the case of hyperfine splitting where the normal energy levels of an excited nucleus are modified in the presence of a magnetic field from its orbiting electrons. The magnetic field will cause each energy level of the nucleus to change slightly and thus create slightly different transitions. This slight change is the called the *experimental energy*. Typically the experimental energy is extremely small compared to the energy level. In order to observe the experimental energy (and thus have an interesting experiment) the linewidth must be narrow enough to observe the shifting or splitting. See Fig. 1c.

The final energy scale in gamma ray fluorescence experiments is the energy lost to a recoiling nucleus. When a nucleus emits a gamma ray, conservation of momentum requires that it recoil in the opposite direction. Likewise, when a nucleus absorbs a gamma ray, it absorbs momentum and begins to move (recoil) in the same direction. The energy associated with the recoiling nucleus is called the *recoil energy*. Since the nuclei we are concerned with will be moving non-relativistically, we may calculated the recoil energy, R, as

$$R = \frac{(p_{nucleus})^2}{2M} = \frac{(p_{photon})^2}{2M} \approx \frac{(E_0)^2}{2Mc^2}$$
(2)

where *M* is the mass of the nucleus and *c* is the speed of light. See Fig. 1d.

- **Exercise 1a:** Derive an expression for Γ using Eq. 1. Assuming an excited nucleus of Fe⁵⁷ has a lifetime of 0.1µs, determine the minimum linewidth. What happens to the linewidth as successively shorter times are used to measure the the energy of the excited state
- **Exercise 1b:** Assume you have a nucleus of Fe⁵⁷ with a transition with energy $E_o = 14.4$ keV. Determine *R* and the ratio of *R* to E_o . Explain

why it was reasonable to approximate $p_{photon} \approx E_o/c$ in the last step of Equation 2. Was it reasonable to assume the nucleus was nonrelativistic? Explain.

Exercise 1c: Assume the hyperfine splitting in a Fe⁵⁷ nucleus is roughly 10⁻⁷ eV. Compare the experimental energy of the hyperfine splitting to linewidth and recoil energy from previous parts. Would you be able to experimentally observe the hyperfine splitting? Explain why or why not.

Introduction: Resonant Absorbtion

In a typical resonance experiment, a gamma ray is emitted by an excited nucleus and then absorbed by a nucleus in the ground state. When emitting the gamma ray, an energy R is lost to the recoiling nucleus. An additional energy R is needed to conserve momentum when exciting the second nucleus. Thus the energy of the gamma ray that is available for exciting a target nucleus is E_o -2R. For resonant absorption to occur, the linewidth of the transition must be large enough to allow an appreciable probability for absorption of this less energetic gamma ray. This leads to the resonance condition

$$2R \le \Gamma \tag{3}$$

Exercise 2: Compare the energy scales from **Exercises 1** and **2** to see if resonant absorption is likely to be observed.

Figure 2. The emitted gamma ray has a peak centered at E_o -R while the absorbing nucleus has a peak centered at E_o +R. Both peaks have the same linewidth Γ . The overlap region is an indication of how probable it is to observe a resonance between the two nuclei. Notice that when $2R \ge \Gamma$ the overlap quickly becomes negligible.

Introduction: Lattices and Phonons

Before Mössbauer's discovery most gamma ray fluorescence experiments were done using a gas. In a gas, every nucleus is essentially acting as a free particle and thus must absorb the entire recoil momentum. As you calculated above, this can make seeing resonance difficult if not impossible. Mössbauer discovered that it was possible to see resonance quite easily using nuclei in the solid state. (See [1] for a more thorough review.) To begin to understand this effect, we must consider what happens when an entire crystal is used as the absorber or emitter. To do this it is necessary to first introduce the concepts of lattices and phonons.

A crystal is comprised of atoms (nuclei) arranged in space at regularly spaced intervals, otherwise known as a *lattice*. See Figure 3. The nuclei on the lattice can be thought of as being connected together by springs. [2] If you hit one of the nuclei, you will cause all of them to vibrate (since they are connected by springs). The lattice vibration is known as a *phonon*. See Figure 3. The phonon is an excitation of the lattice that carries momentum and has an associated energy.

Figure 3.

Just as the energy levels of an atom or a nucleus are quantized, so too are the energy levels of a lattice. Consider the simple model in which each nucleus is attached only to its nearest neighbors by springs. Each nucleus will behave approximately like a harmonic oscillator. You may recall from quantum mechanics that the quantized energy levels of a harmonic oscillator are given by

$$E_n = \hbar \omega (n+1/2) \tag{4}$$

where E_n is the n^{th} energy level and ω is the angular frequency of oscillation. Notice the important fact that the ground state energy (n=0) is non-zero. This means that to make that lattice vibrate (or create phonons) the lattice must absorb at least $\hbar \omega / 2$ in energy.

Exercise 3: The minimum phonon energy corresponds to the Debye temperature, Θ_D . Determine the phonon energy using the equation $E_{phonon} = k_b \Theta_D$ with k_b being the Boltzmann constant and using a table from [2] or the CRC.

Introduction: Mössbauer's Discovery

Mössbauer used solid state absorbers and emitters in gamma ray fluorescence experiments. The recoil energy from single nuclear decay events in a lattice can be too low to excite phonons, which means that for the purpose of recoiling, the decaying nucleus is effectively rigidly bound to all the other nuclei in its crystal lattice. Since the recoil momentum must be imparted to the entire lattice, not just a single nucleus, the kinetic energy that goes into the recoil drops dramatically. This recoilless absorbtion and emission of gamma rays is known as the *Mössbauer effect*.

Mössbauer's discovery has had far-reaching consequences because it has made available electromagnetic radiation (gamma rays) whose frequency is more precisely defined than any other known to date. Energy resolution better than one part in 10^{12} has been achieved with recoil-free gamma rays.

The isotope with the strongest recoiless resonant absorption is Fe^{57} . Figure 4 shows the decay of Fe^{57} from its parent Co^{57} . Of all the excited Fe^{57} nuclei, about 10% will emit a 14.4 keV gamma ray via a magnetic dipole transition from the metastable I = 3/2 state to the I = 1/2 ground state (I is the nuclear spin). The ratio of recoil-free 14.4 keV photons to all the 14.4 keV photons emitted is f, the recoil-free fraction of the source. f varies with the properties of the solid and decreases monotonically with increasing temperature.



- **Figure 4.** Energy level scheme of Fe⁵⁷. Mössbauer spectroscopy involves the 14.4 keV transition. Intensities are given in % of decays.
- **Exercise 4a:** What is the minimum recoil energy needed to produce a phonon (use the result from **Exercise 3**)?
- **Exercise 4b:** Consider the Fe^{57} transition discussed above. Are the linewidth and recoil energy sufficient to excite a phonon?
- **Exercise 4c:** Assume the recoil momentum is imparted to the entire lattice of N nuclei instead of a single nucleus. What is the recoil momentum of the lattice as a whole? What is the recoil energy?

Introduction: Level Splitting

When a nucleus is placed in a magnetic field its energy levels are split. If the magnetic field is due to the electrons orbiting the nucleus, the effect is called hyperfine splitting. Hyperfine splitting is another form of the Zeeman effect ([3], ch. 7). Since spin of the nucleus gives it a magnetic moment, the energy of transitions is shifted by an amount

$$\Delta E = -\mu H / I \tag{5}$$

where μ is the magnetic moment of the nucleus, *H* is the magnetic field splitting the levels, and *I* is the spin of the nucleus. The minus sign reflects that fact that a magnetic moment aligned with the field is more energetically favorable. Recall that the spin quantum number, m_j , ranges from -I to I (2*I*+1 total values). The energy level splitting for a nucleus with a particular m_i is just

$$\Delta E = -\mu H m_j / I \tag{6}$$

The Fe⁵⁷ nucleus used in this experiment has spin 3/2 and spin 1/2 states. The transition between these two states is the previously mentioned 14.4 keV transition with a 0.1µs lifetime. In the presence of a magnetic field, both the excited state and ground state energy levels are split. Only a subset of all imaginable transitions between the split states are allowed [3].

- **Think!** What is the physical reason that some transitions are allowed and others are forbidden?
- **Exercise 5a:** Calculate the energy shift for I=1/2 using Eq. 6. Assume the magnetic field at the nucleus is 10^5 Gauss and the magnetic moment of the ground state (I=1/2) is $0.093\mu_n = 2.93 \cdot 10^{-12}$ eV/Gauss.
- **Exercise 5b:** The unperturbed energy of transition from I=3/2 to I=1/2 is 14.4 keV. Calculate the shift in the $m_j = -3/2$ energy level. Assume $\mu_1 = -\mu_o$ is the magnetic moment of the excited state (I=3/2). Calculate the transition energy between states $m_j = -3/2$ and $m_j = +1/2$. Is the linewidth of the 14.4 keV transition narrow enough to see the splitting?
- **Exercise 5c:** Calculate the energy difference between the $m_j = -3/2$ to $m_j = +1/2$ transition and the $m_j = -1/2$ to $m_j = +1/2$ transition in terms of the magnetic moment of the first excited state μ_i



Figure 5. The transition from I=3/2 to I=1/2 gets split into six possible transitions. Notice that the I=3/2 levels are flipped upside down. This is because the magnetic moment of the excited state carries a negative sign. See figure 6 from [1].

Introduction: Doppler Shift

An experimental shift similar in scale to the hyperfine splitting is that of the Doppler shift. If a nucleus is moving when it decays, the energy of the emitted gamma rays will be shifted. If it is moving toward an absorber, higher energy gamma rays will be seen at the absorber. If it is moving away from the absorber, lower energy gamma rays will be seen. This Doppler shift changes the gamma ray energy by an amount

$$\Delta E = \frac{vE_0}{c} \tag{7}$$

where v is the speed of the source. (For a derivation, see [3].)

Exercise 6: Estimate the velocity needed to counteract the hyperfine splitting in Fe^{57} using the result of **Exercise 5**.

Materials and Methods

The Mössbauer effect allows for very precise measurements of gamma ray resonance. In combination with the Doppler shift, it can be used to observe the hyperfine splitting of the ⁵⁷Fe nucleus and thus determine the internal field of the ⁵⁷Fe nuclei. A measurement of the linewidth can also be made.

In our experiment, a FeCo nucleus decays into an excited state of the Fe⁵⁷ nucleus (Figure 5). Since FeCo is not ferromagnetic, the decaying nucleus does not experience any Zeeman splitting and emits 14.4 keV gamma rays. The emitted gamma rays are incident on an absorber with roughly 2.2% abundance of ⁵⁷Fe nuclei. This concentration is sufficient that any absorbing nucleus experiences Zeeman splitting due to the magnetic field from surrounding ⁵⁷Fe atoms. If the source is at rest relative to the absorber, the splitting of energy levels in the absorber prevents resonant absorbtion. If the relative velocity of the source in the direction of the absorber increases, Doppler shifted gamma rays can

be made to match resonance conditions with the split levels in the absorber. By changing the velocity of the source, resonance conditions can be reached for each of the different transitions in the absorber. When in resonance, the absorber will absorb much more of the gamma rays. The shape of the excess absorbtion peaks may be compared to theoretical estimates and the natural linewidth may be determined. Our experimental setup is similar to the one discussed in [3].

The experimental apparatus is drawn in Figure 6. A radioactive FeCo source is mounted on a vibrating shaft that moves in the direction of a Geiger Mueller tube (detector). An absorber placed between the source and the detector may limit the number of gamma rays that reach the detector. Several pieces of electronic equipment are use to control the motion of the source and correlate it with the signal from the detector.

Materials and Methods: *Devices*

Constant Acceleration Drive

You will use a Constant Acceleration Drive (CAD) to scan through the different velocities. The constant acceleration provides for a linear curve when velocity is plotted as a function of time. The source begins with a minimum (negative) velocity and accelerates to a maximum (positive) velocity. Upon reaching a maximum velocity the cycle repeats itself. On average there will be an equal number of gamma rays emitted at each possible velocity. By counting the number of a gamma rays one can determine the number of absorbtions that occur at each velocity. A relative excess of absorbtions indicates resonant absorbtion.



- Figure 6. Schematic of the experimental setup.
- **Exercise 7a:** Sketch the acceleration, velocity, and position versus time using the information from the paragraph above.
- **Exercise 7b:** If the counting device you are using has about 250 velocity increments, approximate how many counts will be observed at each velocity increment if 10,000 transition are observed in a single run. What is the error in a single channel(assume error goes as \sqrt{N})?
- **Exercise 7c:** Approximate how many runs are need to achieve 1% accuracy per

channel?

Exercise 7c: If the decay time of the Fe^{57} nucleus is 0.1 µs, determine the minimum time it would take to get 1% accuracy in each channel? Explain possible reasons why the real time to perform this many runs is actually much greater (hint: where does the ⁵⁷Fe nucleus come from).

Multichannel Analyzer

To keep track of the number of gamma rays detected at each velocity you will use a Multichannel Analyzer (MCA). The MCA has several hundred independent channels with which to record counts from the detector. The active channel of the MCA is triggered by the voltage applied to the CAD. If the CAD is at its maximum velocity, a count from the detector will be recorded in the lowest channel of the MCA (channel 0). If the CAD is at its minimum velocity, the MCA will record counts in the highest channel (channel 255). Each channel in between inactive at an intermediate velocity. Since each velocity corresponds to a small energy change the energy of the emitted gamma ray, the absorbtion spectrum above and below the Fe⁵⁷ transition can be plotted.

Exercise 8: Make an educated guess as to the range of velocities you will be using. Knowing that the velocity changes linearly with time, you may assume that each channel will be equally spaced in velocity. Determine the width of the velocity bins (*i.e.*, how many mm/s are read into a single channel). What is the corresponding energy resolution? Would this resolution be adequate to see the Zeeman splitting (see **Exercise 5**)?

Materials and Methods: *Procedure*

- I. Turn on
 - A. MCA
 - B. Main power switch for equipment rack
 - C. Printer (before computer)
 - D. Computer and monitor

II. Settings

- A. Constant Acceleration Drive
 - 1. amp adj 3.1
 - 2. freq trim on mark
 - 3. gain on mark
 - 4. 200 µsec
 - 5. start
 - 6. int
- B. Oscilloscope (2213A)
 - 1. 60amp-DC, 5V/div (vert.)
 - 2. 2A60 amp-DC, 0.05 V/Div (horiz.)
- C. Filter Amplifier (Ortec 440A)
 - 1. gain-high, 32
 - 2. fine gain 0
 - 3. shaping time 1μ sec
 - 4. outputs unipolar
 - 5. input pos
- D. Power Supply
 - 1. output 2000 volts
 - 2. polarity pos
- III. Insert Source
 - A. Use Pb gloves and tongs
 - B. Turn off constant acceleration drive
 - C. Screw on source
 - D. Turn on constant acceleration drive
 - E. Adjust gain and frequency to just below high frequency tone
- IV. MCA
 - A. Read manual, pp 1-28
 - B. When MCA is turned on, several self tests are conducted
 - 1. A BOX around a word indicates a key to be pressed
 - 2. Press CONT to finish self tests.
 - C. Clear memory: $\boxed{0} \frac{\text{GOES}}{\text{INTO}}$ ALL
 - D. $\frac{AMP}{DIRECT}$ until <u>DIR</u> is on in bottom right corner. <u>TERM</u>

V. PHA

- SET UP , PHA, CONT (add), CONT set on Q1, CONT set on <u>NONE</u>, TERM
- B. DISPLAY Q1
- C. Remove 57 Fe Foil from Geiger-Muller tube
- D. RUN, PHA, set vert=log or vert=2k
- E. Select 14.4 keV line
 - 1. Four peaks are on PHA
 - 2. 6keV gamma rays are stopped by a magazine
 - 3. An aluminum plate will stop 14.4 keV gamma rays
 - 4. Use LLD to eliminate 6keV and below
 - 5. Use ULD (small screwdriver) to eliminate upper peaks.
 - 6. TERM

VI. Multichannel scaler

- A. Put ⁵⁷ foil on Geiger-Muller tube
- B. $\frac{\text{SET}}{\text{UP}}$, MCS, CONT (add), CONT set on Q1, CONT set sw=9999, CONT set
- on <u>EXT</u>, <u>TERM</u> C. <u>DISPLAY</u> Q1, \boxed{O} <u>GOES</u> <u>ALL</u>
- D. Single run of MCS is 9999 sweeps.
 - You need ≥ 4 sweeps for a good histogram
 - 1. For single run: RUN MCS
 - 2. For several runs:
 - a. PROG, 1, EDIT, RUN, MCS, EXIT EDIT
 - b. DISPLAY Q1
 - c. To run: RUN, press a number for # of runs, PROG, 1
- VII. Input-Output
 - SET UP, I/O, CONT (ser), CONT (1), CONT (out), CONT (data), CONT (set A. for Q1), TERM
 - B. baud rate: OPTIONS, $\frac{CONT}{LIST}$, BAUD (increment baud rate until BAUD=19200) (keep pushing BAUD), DELAY (off), TERM
- VIII. Transfer data to computer
 - A. Type MOSSBAUER, enter
 - B. Number of resonances = 6
 - C. RUN, I/O
 - D. set scale: use <u>PRINT SCREEN</u> to print graph
 - E. any key Y/N
 - F. Record computer information

G. Print data - use PRINT SCREEN

Resonant Absorption: Analysis

- 1. Measure the energy shifts for all transitions and deduce the magnetic moment of the first excited state of Fe^{57} (*I*=3/2) and the size of the internal magnetic field felt by the ⁵⁷Fe nucleus.
- 2. Calculate the error in your data with and without including the background counts. Assume the number of transitions at each energy has a Gaussian distribution. (See [4], section 2.3) (hint: what do μ and σ mean when you are considering the distribution of counts?). Do your error estimates seem reasonable based on the appearance of your plot (especially the noise level between absorbtion peaks)?
- 3. The distribution of counts is not actually Gaussian but really obeys a Lorentzian distribution (also known as the Breit –Wigner Distribution in nuclear physics). (See [4], section 2.4.) Does it make sense to give error estimates for the magnetic moment of the first excited state and the internal field? Describe how to use a fitting routine to get true error estimates from the data. You will definitely want to consult chapters 6 an 8 of [4]. What problems would you encounter in performing a least squares fit on the data? Explain.
- 4. Assuming that both the split and unsplit levels have the same natural linewidth. Estimate the linewidth. Don't forget to account for the fact that both the emitter and absorber have a linewidth (how does this affect the width of the absorbtion peaks? See [1]). Compare your result with the accepted value. If your result deviates from the theoretical result, discuss why.
- 5. Describe how a transition actually gets registered as a count. Explain how the gamma ray passing through the absorber is converted into a count on the MCA. Explain the purpose of setting the window on the PHA (you may find it useful to read the section in the MCA manual entitled ADC Setup). Explain how the MCA decides which channel to record a count. Be specific about how the start of each MCA sweep is determined. What would happen if the channel advance toggle was changed to 400 µs?
- 6. How would changing the LLD and ULD affect the data/experiment? Would it be advantageous to use a smoothing algorithm on the data? Which part of the analysis (1-5) would most likely improve when using smoothing?

References

[1] H. Lustig, Trans. of the New York Academy of Sciences 23, 375 (1961).

[2] C. Kittel, Introduction to Solid State Physics (Wiley, New York, 1996).

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[4] P. R. Bevington and D. K. Robinson, *Data Reduction and Error Analysis for The Physical Sciences* (McGraw-Hill, New York, 1991).