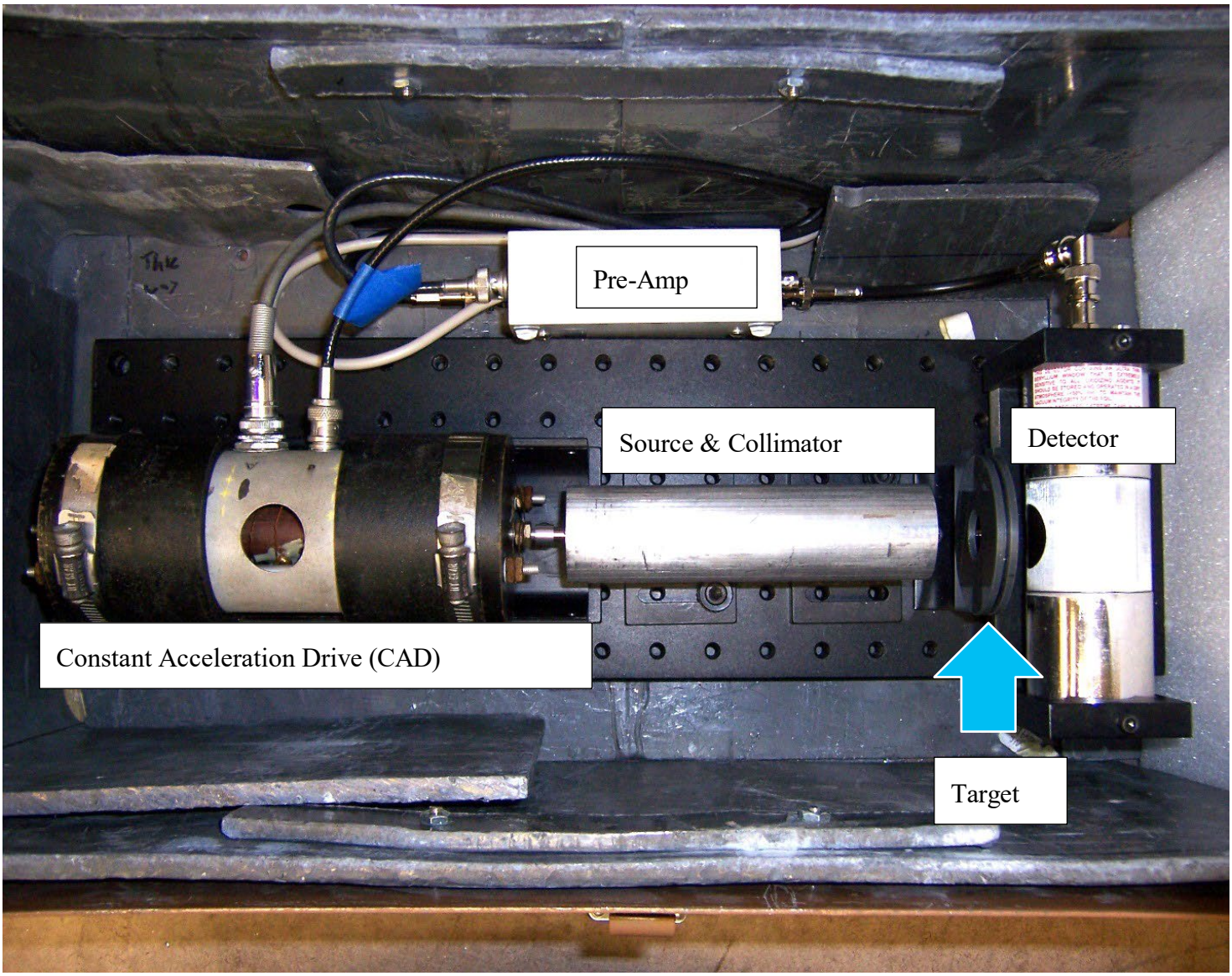
The Mössbauer Effect

UCSB Physics Department, October 22, 2024

What is in the box?



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 ^{57}Fe 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 the nucleus in its ground state. When a nucleus decays from an excited state, the excitation energy is often radiated away as a gamma ray. Typically, the energies of such gamma rays are less than 1 MeV. [1]

Because of quantum mechanical effects, there is a small spread in the possible energy values of the emitted gamma ray. The uncertainty principle states that

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

where $\hbar$ is Planck's constant divided by 2π, 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. Since the excited state cannot exist for longer than the lifetime, however, τ gives a lower bound on the energy spread ΔE_0. This energy spread is called the ***linewidth*** and is denoted Γ. Experimentally one determines it by measuring the width of an energy peak at its half maximum. The energy scale for Γ is usually many orders of magnitude smaller than E_0. [1]

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 perturbed, for example, by the presence of an external magnetic field. This perturbation removes the degeneracy of the components of any levels that have more than one, causing them to split into multiple levels that have different energies. This gives rise to multiple transitions that result in absorption or emission of amounts of energy that are slightly different from the energy that would be absorbed or emitted by the single transition that would occur between two unperturbed (and therefore unsplit) levels. This slight difference in absorbed or emitted energy is called the ***experimental energy***. Typically, the experimental energy is extremely small compared to the absorbed or emitted energy. In order for one to observe the experimental energy (and thus have an interesting experiment) the linewidth must be narrow enough to observe the shifting or splitting.

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 calculate the recoil energy, R, as

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

where M is the mass of the nucleus and c is the speed of light.

Exercise 1a: Derive an expression for Γ by using Eq. 1. Assuming an excited nucleus of ^{57}Fe has a lifetime of 0.1 μs, determine the minimum linewidth. What happens to the linewidth as successively shorter times are used to measure the (relative) energy of the excited state?

Exercise 1b: Assume you have a nucleus of ^{57}Fe 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 use the approximation $p_{photon} \approx E_o/c$ in the last step of Equation 2. Was it reasonable to assume the nucleus was non-relativistic? Explain.

Exercise 1c: Assume the hyperfine splitting in a ^{57}Fe nucleus is roughly 10^{-7} eV. Compare the experimental energy of the hyperfine splitting to the linewidth and recoil energy from previous parts. Would you be able to experimentally observe the hyperfine splitting? Explain why or why not.

Introduction: Resonant Absorption

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, the nucleus loses an energy R as it recoils. An additional energy R is needed to conserve momentum when the second nucleus recoils as it absorbs the gamma ray. Thus, the energy of the gamma ray that is available for exciting a target nucleus is $E_0 - 2R$. 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

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

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

Introduction:
Lattices and Phonons

Before Mössbauer's discovery, most gamma ray fluorescence experiments were done on gaseous samples. 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 by 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 first to introduce the concepts of lattices and phonons.

A crystal comprises atoms (nuclei) arranged in space at regular intervals, otherwise known as a ***lattice***. You can think of the nuclei in the lattice 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). This lattice vibration is known as a ***phonon***. The phonon is an excitation of the lattice that carries momentum and has an associated energy.

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 as 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.

Exercise 3: The minimum phonon energy corresponds to the Debye temperature, Θ_D. Determine the phonon energy by 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 absorption and emission of gamma rays is known as the ***Mössbauer effect***.

Mössbauer's discovery has had far-reaching consequences. Because of the wide variety of extremely precise energy measurements it makes possible, Mössbauer was awarded a Nobel Prize in 1961. Such measurements have yielded a wealth of information on nuclear structure and crystal structure, and have even been used to verify predictions of relativity theory. For example, with a vertically oriented, 22.5-meter-high Mössbauer spectrometer, scientists measured the gravitational red shift of gamma rays. They measured this extremely small shift of about 2 parts in 10^{15} with an accuracy of better than 10%, and their measurement agreed with the prediction of Einstein's theory of general relativity within that accuracy. [3]

The isotope with the strongest recoilless resonant absorption is ^{57}Fe. Figure 1 shows the decay of ^{57}Fe from its parent ^{57}Co. Of all the excited ^{57}Fe 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.

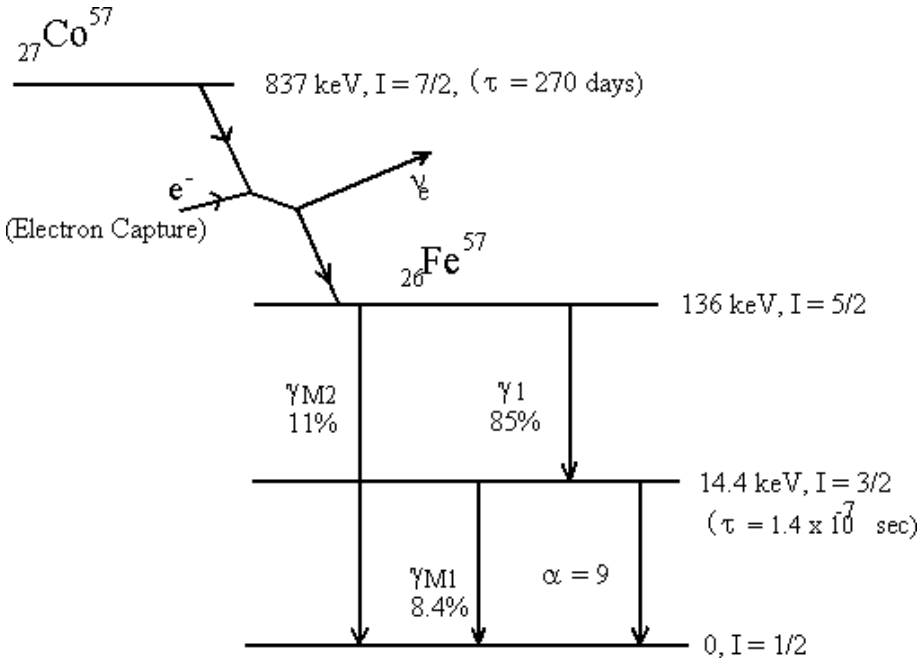


Figure 1. Energy level scheme of ^{57}Fe. Mössbauer spectroscopy involves the 14.4-keV transition. Intensities are given in % of decays.

Exercise 4a: What is the minimum recoil energy needed to produce a phonon (use the result from **Exercise 3**)?

Exercise 4b: Consider the ^{57}Fe 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. This magnetic field may be due to the magnetic dipole fields of neighboring nuclei, or to electric quadrupole fields of electrons orbiting the nucleus or in neighboring atoms. This effect is called hyperfine splitting, and is a form of the Zeeman effect ([4], ch. 7). It arises from coupling of the nuclear magnetic moment with the external field. Because of the cubic symmetry of the lattice in which the ^{57}Fe sits, the electric quadrupole field from either its electrons or those of neighboring atoms does not lift the degeneracy of the nuclear spin states, and you will observe only the Zeeman effect from the magnetic field due to neighboring nuclei. [5] Since spin of the nucleus gives it a magnetic moment, the energy of a particular state (level) 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 the fact that alignment of a magnetic moment with the field is more energetically favorable than alignment antiparallel to it. Recall that the spin quantum number, m_j, ranges from $-I$ to I ($2I + 1$ total values). The energy level shift for a nucleus with a particular m_j is just

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

The ^{57}Fe nucleus has spin 3/2 and spin 1/2 states. The upper, spin 3/2 state has a 0.1-μs lifetime. The transition between these two states is the previously mentioned 14.4-keV transition. 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 is allowed [4]. Figure 2, on the next page, shows the levels that result from this splitting.

Think! What is the physical reason that some transitions are allowed and others are forbidden?

Exercise 5a: Determine the magnetic moment of ^{57}Fe in the ground state ($I = 1/2$). Go to this website, https://www-nds.iaea.org/nuclearmoments/, and select the ^{57}Fe isotope. The units for the magnetic moment are given in nuclear magnetons, μ_N (nm), meaning that you need to multiply the number in the table by μ_N. Note that $\mu_N = \frac{e\hbar}{2m_p}$ where e is the elementary charge and m_p is the mass of the proton. Give your answer for the magnetic moment in eV/gauss.

Exercise 5b: Calculate the energy shift for each level of the $I = 1/2$ state ($m_j = -1/2$ and $m_j = +1/2$) by using Eq. 6. Assume the magnetic field at the nucleus is 10^5 gauss.

Exercise 5c: The energy of the transition between the unperturbed $I = 3/2$ and $I = 1/2$ states is 14.4 keV. Assuming that $\mu_1 = -\mu_o$ is the magnetic moment of the excited state ($I = 3/2$), calculate the shift for the $m_j = -3/2$ and $m_j = -1/2$ levels of this state. From these shifts, calculate the energy difference between these two levels, to both of which the transition from the $m_j = -1/2$ level in the ground state is allowed. Is the linewidth of the 14.4-keV transition narrow enough to see this splitting?

Exercise 5d: Now use μ_1 given at the link above to calculate the shift for each level in the excited state, and calculate the energy difference between the $m_j = -3/2$ and $m_j = -1/2$ levels.

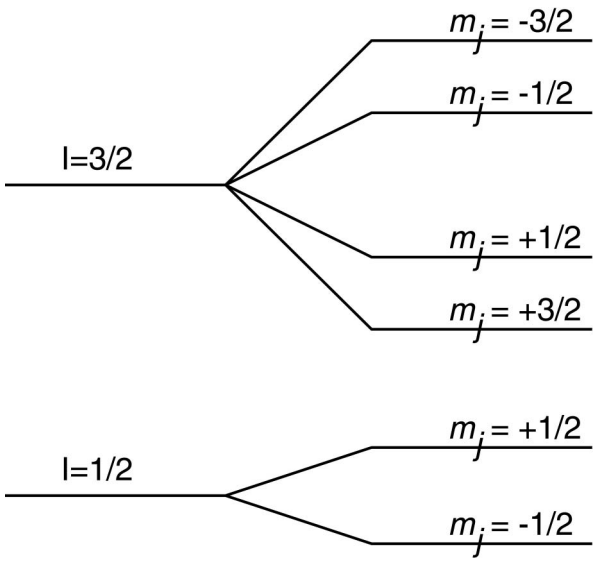


Figure 2. The transition from I=3/2 to I=1/2 gets split into six possible transitions. Notice that the I=3/2 levels are flipped upside down. This is because the magnetic moment of the excited state carries a negative sign. See figure 3 from [1].

Introduction: Doppler Shift

One can produce an energy shift similar in scale to the hyperfine splitting by means of the Doppler effect. If a nucleus is moving when it decays, the energy of the emitted gamma rays is shifted. If it is moving toward an absorber, the gamma rays that reach the absorber are higher in energy than those emitted by the stationary nucleus. If it is moving away from the absorber, the absorber receives gamma rays that are lower in energy than those emitted by the stationary nucleus. 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 [4].)

Exercise 6: Use the result of **Exercise 5** to estimate the velocity needed to match the hyperfine splitting in ^{57}Fe.

Materials and Methods

The Mössbauer effect allows for very precise measurements of gamma ray resonance. In combination with the Doppler shift, one can use it to measure the hyperfine

splitting of the ^{57}Fe nuclear states and thus determine the magnetic field due to neighboring ^{56}Fe nuclei. A measurement of the linewidth can also be made.

In our experiment, a ^{57}Co nucleus decays into an excited state of the ^{57}Fe nucleus (Figure 2). The environment in which the ^{57}Co sits is such that the decaying nucleus does not experience any Zeeman splitting, and the excited ^{57}Fe emits 14.4-keV gamma rays. The emitted gamma rays are incident on an absorber with roughly 2.2% abundance of ^{57}Fe nuclei. (This is the natural abundance of this isotope.) Most of the foil – about 92% – comprises ^{56}Fe atoms, whose nucleus has a significantly larger magnetic moment than that of ^{57}Fe. As noted above, the crystal structure of the foil prevents electric quadrupole interaction between an ^{57}Fe nucleus and any of the surrounding electrons. The ^{57}Fe nuclei in the foil thus experience Zeeman splitting of their energy levels due to the magnetic field from the nuclei of the surrounding ^{56}Fe atoms.

If the source is at rest relative to the target, the splitting of energy levels in the target prevents resonant absorption. As noted above, if we move the source toward the target, we increase the energy of the gamma rays it receives, and if we move it away from the target we decrease the energy of the gamma rays it receives. If we vary the speed of the source, then, we can vary the energy of the gamma rays reaching the target, so that resonance occurs for each allowed transition in turn. When this happens, the target absorbs many more gamma rays than it does when their energy is not resonant with a transition, and we see a dip in the number of counts from the detector. As we vary the velocity of the source, then, we observe a series of peaks, each of which corresponds to an allowed transition. You can compare the shape of these excess absorption peaks to theoretical estimates, and you can determine the natural linewidth. Our experimental setup is similar to the one discussed in [4]. The 2.2% concentration of ^{57}Fe atoms in the foil is sufficient to provide measurable absorption of gamma rays when resonance occurs.

The experimental apparatus is drawn in Figure 3. A radioactive ^{57}Co source is mounted on a vibrating shaft that moves in the direction of a Geiger-Mueller tube (detector). You may limit the number of gamma rays that reach the detector by placing an absorber between it and the source. Several pieces of electronic equipment are used 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 source velocities. The constant acceleration provides for a linear graph when you plot velocity as a function of time. The source begins with a minimum (greatest negative) velocity and accelerates to a maximum (greatest positive) velocity. Upon reaching the maximum velocity it ramps back down to the original minimum velocity. It takes 200 ms to complete this cycle. The arbitrary waveform generator is triggered by the MCA at the beginning of each pass to start a new cycle. Because there are 1024 channels, each with a dwell time of 200 μs, a single pass of the MCA takes 204.8 ms to complete. Therefore, there is a dead time of 4.8 ms (CAD not moving) for each pass.

On average there will be an equal number of gamma rays emitted at each possible velocity. By counting the number of gamma rays you can determine the number of absorptions that occur at each velocity. A relative excess of absorptions indicates resonant absorption.

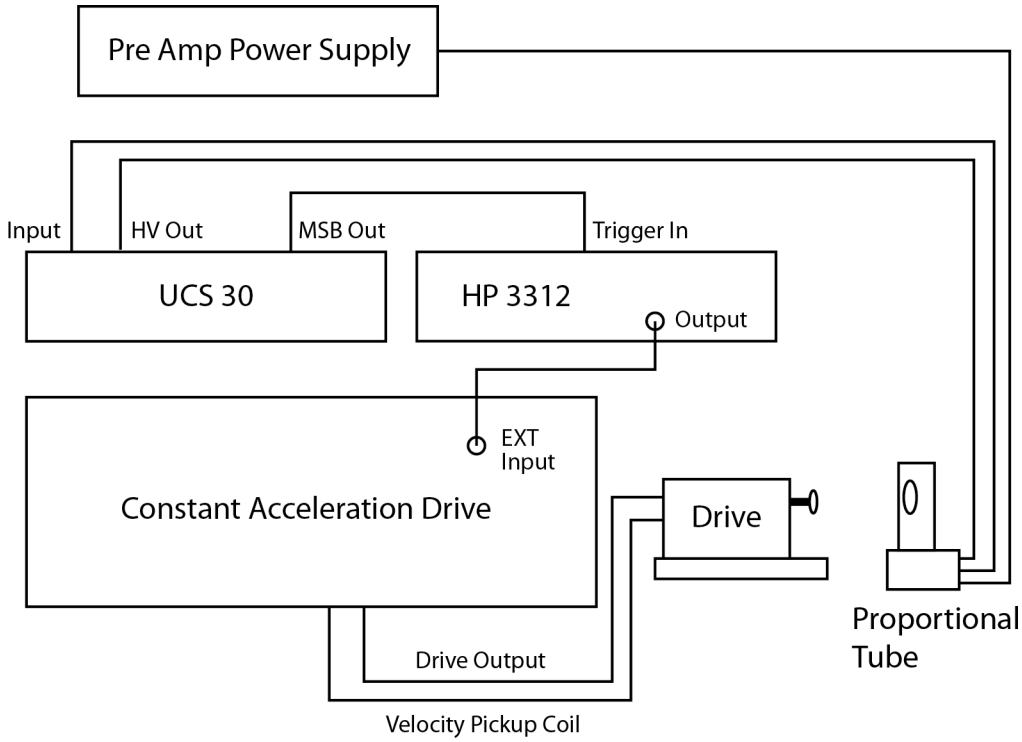


Figure 3. Schematic of the experiment setup.

Exercise 7a: Sketch the acceleration, velocity and position versus time, based on the information from the paragraph above.

Exercise 7b: If the counting device you are using has 250 velocity increments, estimate how many counts you will observe at each velocity increment if you observe 10,000 transitions in a single run. What is the error in a single channel (assume that the error goes as $\sqrt{N}$)?

Exercise 7c: Approximately how many runs are need to achieve 1% accuracy per channel?

Exercise 7c: If the decay time of the ^{57}Fe nucleus is 0.1 μs, determine the minimum time it would take to get 1% accuracy in each channel? Explain possible reasons for which the real time to perform this many runs is actually much greater (hint: where does the ^{57}Fe 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 MCA automatically passes through each channel for the specified dwell time. If the CAD is at its minimum velocity, a count from the detector will be recorded in the lowest channel of the MCA (channel 0). If the CAD is at its maximum velocity, the MCA will record counts near the middle channel (channel 500, not 512; remember the velocity wave is a 5Hz triangle wave, ***but see***

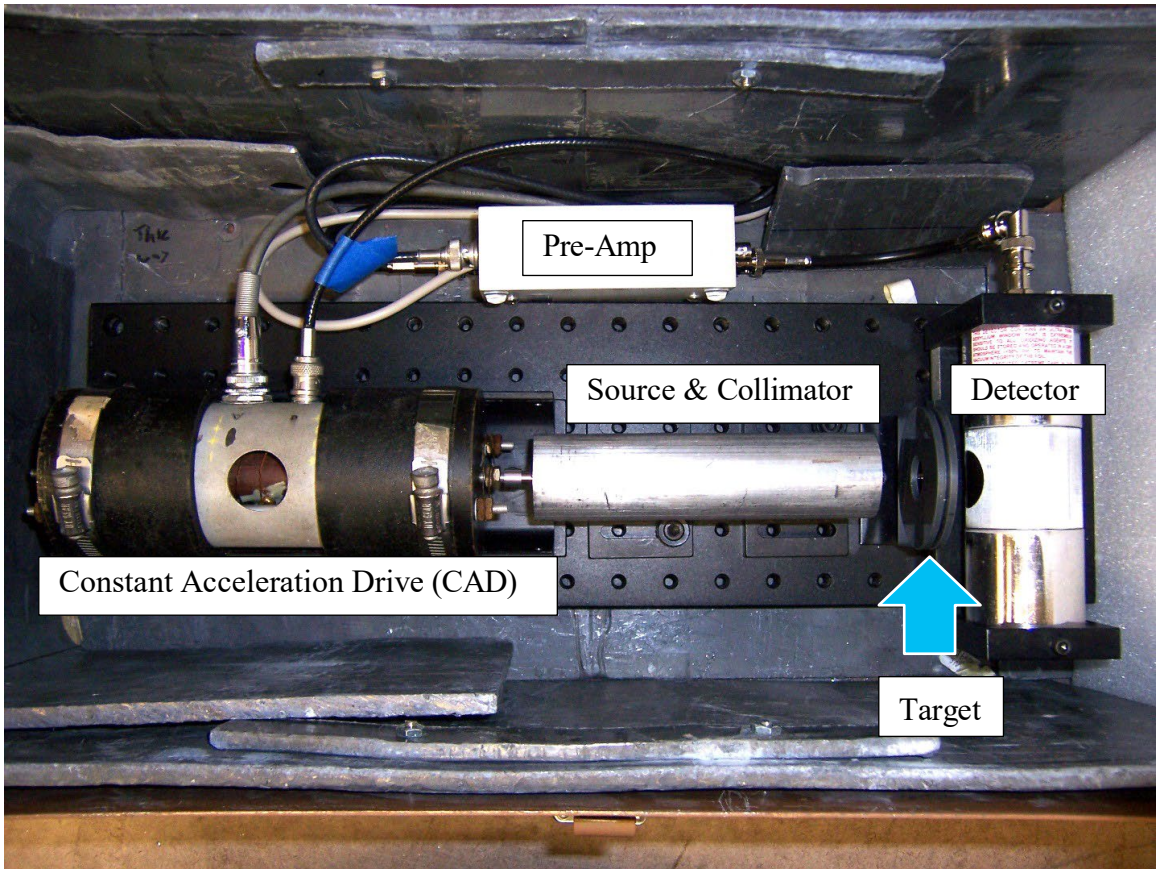
Below!). Since each velocity corresponds to a small change in the energy of the emitted gamma ray, you will be able to obtain the absorption spectrum above and below the ^{57}Fe transition (between the unsplit levels).

Important note: Because of the way the UCS 30 works, the trigger (channel 0) appears at channel 512 (511 in the .csv file). The right-hand spectrum (channels 512 to 1011 (511 to 1010 in the .csv file) is the one to analyze. The left-hand spectrum is from the ramp back down from maximum velocity to minimum velocity, so it is the reverse of the one on the right. (The spectrum wraps around.)

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 the channels 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**)?

What's in the box?

Here is a photograph of the inside of the steel box with the lead shielding removed.



Materials and Methods:
Procedure

1) Turn on the main power to the instrument rack.
2) The settings for the instruments in the rack are shown below for reference, but should not need changing. **Please note that these settings determine how the CAD operates. You will not use the CAD until you have run the spectrometer in pulse height analysis (PHA) mode to look at the ^{57}Co gamma ray spectrum. DO NOT adjust anything on the green CAD drive panel or the arbitrary waveform generator without consulting a TA or Lab Manager.**

   CAD Drive
   - EXT Input
   - Gain should be pointing to top line

   Arbitrary Waveform Generator
   - Waveform: Triangle
   - Frequency: 5 Hz
   - Amplitude: 8 V Peak to Peak
   - Phase Shift: -90 degrees
   - Burst Mode (one cycle) with External Trigger

3) Locate and start the UCS 30 program. This communicates with the MCA you will be using.
4) First you'll want to look at the spectrum of the ^{57}Co source. Make sure there are no absorbers between it and the proportional counter tube.
5) In the UCS 30 program click on the AMP/HV/ADC settings button  at the top of the toolbar. This dialog box allows you to control the high voltage supply (powering the proportional counter) inside the analyzer, as well as some gain and discriminator settings. Make sure the settings appear as shown in Figure 4.
6) Now you are ready to look at the ^{57}Co Spectrum. Under the Mode menu select **PHA (Amp In)**. Then click "Go." See Figure 5
7) After a minute or two you should see something similar to the image below (Figure 6). You can use the buttons to change the x scale; you want to see only channels 53-1024.

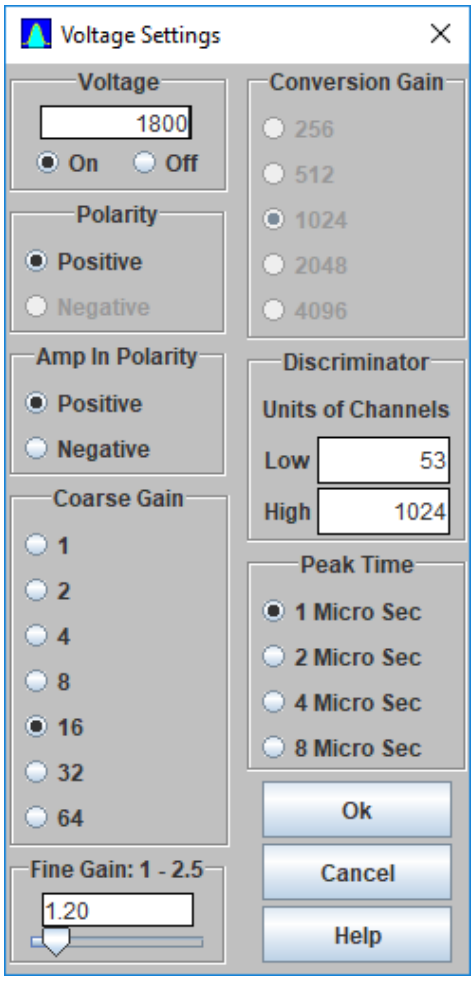


Figure 4

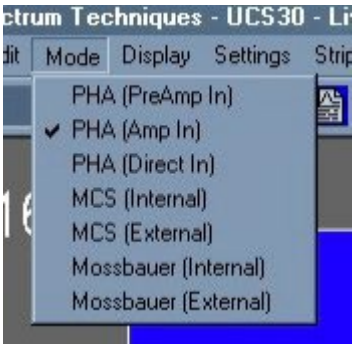


Figure 5

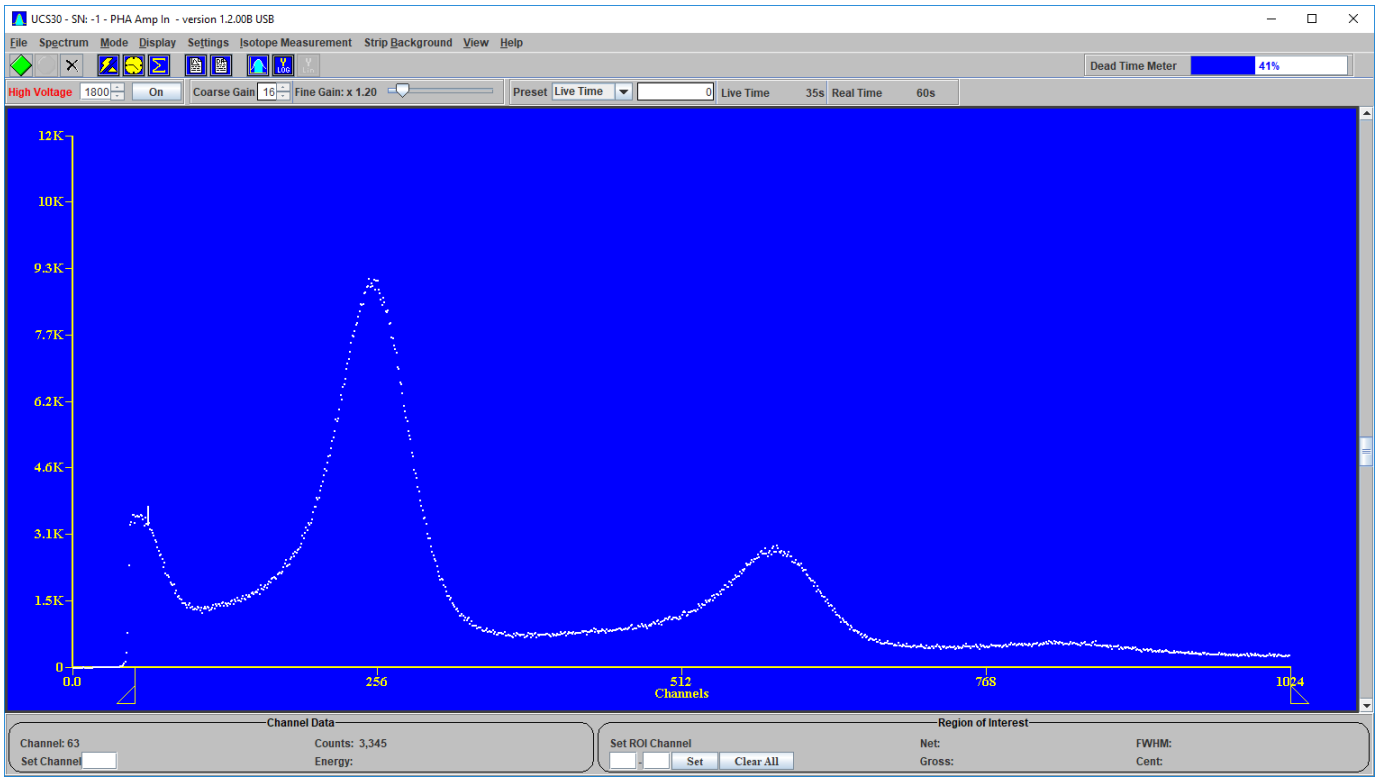


Figure 6

8) Now you need to identify the 14.4-keV peak. You are interested only in this energy, and do not want to register counts outside of it once you change to Mössbauer mode. You should see a few peaks from the ^{57}Co, as in Figure 6. A thin aluminum absorber will block energies **below** 14.4 keV. Place the aluminum sheet between the collimator and the detector to deduce the location of the 14.4-keV peak. It may be helpful to continually erase the spectrum so you can see it build up.
9) Once you have settled on which peak is the 14.4-keV peak, drag the small triangles at the far sides of the x-axis to allow only the 14.4-keV peak to go through the discriminator. These triangles control the onboard discriminator, which will reject measurements whose values lie outside of the selected region.
10) Once the discriminator is set, you are ready to begin the main data collection. Stop the PHA acquisition. Remove the aluminum sheet, and place the ^{57}Fe-enriched foil in between the source and detector.
11) Under the Mode menu select "Internal Mössbauer."
12) Under the Settings Menu, select Mössbauer Settings. **You must set Mössbauer settings through this menu and not through the main screen in order for them to take effect.**
13) Set the Dwell Time to 200 μs and the Number of Passes to 72000. You can stop the acquisition at any time, so you can just wait until you see a good spectrum before stopping. (If you leave the number of passes set to zero, data collection continues for as many passes occur before you stop it.)
14) Clear the display and press Go. Data collection will begin. The traces on the oscilloscope, which shows the trigger generated by the UCS 30, and the triangle wave produced by the arbitrary waveform generator, should look as shown in figure 7 below.

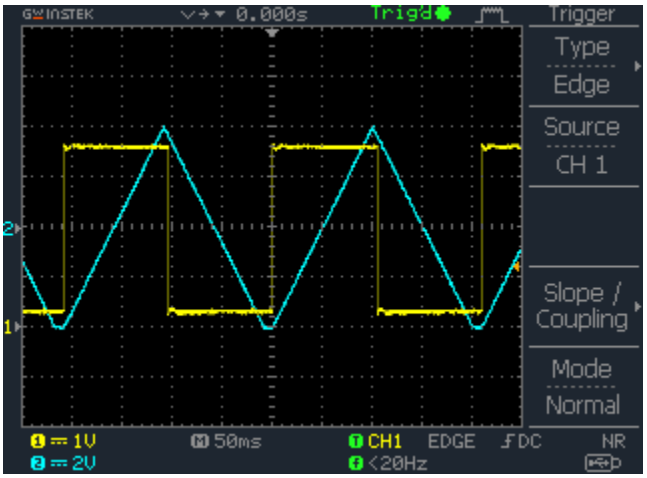


Figure 7

15) After a while (30 minutes to three hours, depending upon the activity of the source) you should start to see the absorption spectrum appear as shown below.

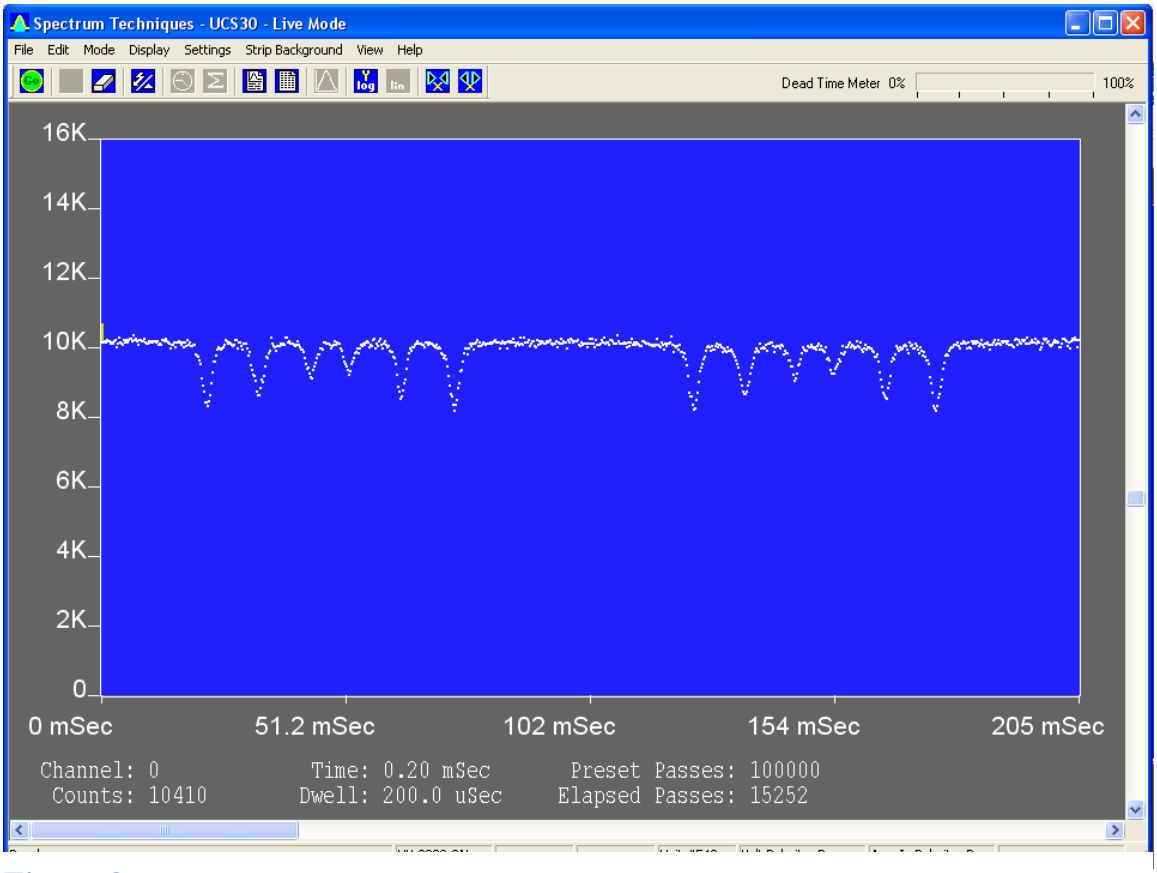


Figure 8

The motion of the CAD has already been calibrated with an interferometer. Analysis showed that the CAD ramps from -9.59 mm/s to +10.22 mm/s and back down in 200 ms. The raw data file consisting of a 1-second recording of the time series output of the interferometer is available for independent analysis.

Resonant Absorption: *Analysis*

1. Measure the energy shifts for all transitions and deduce the magnetic moment of the first excited state of ^{57}Fe (I = 3/2) and the size of the internal magnetic field felt by the ^{57}Fe 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 [6], 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 absorption 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 [6], 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 and 8 of [6]. What problems would you encounter in performing a least squares fit on the data? Explain.

4. Assuming that transitions between 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 absorption 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 titled, "ADC Setup"). Explain how the MCA decides in which channel to record a count. Be specific about how the start of each MCA sweep is determined. What would happen if the dwell time were 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 be improved by the use of smoothing?

References

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[2] C. Kittel. *Introduction to Solid State Physics* (New York: John Wiley & Sons, 1996).

[3] Robert Eisberg and Robert Resnick. *Quantum Physics of Atoms, Molecules, Solids, Nuclei, and Particles* (New York: John Wiley & Sons, 1974), pp. 634-636.

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