Experiment 28

THE MÖSSBAUER EFFECT

THE MÖSSBAUER EFFECT

INTRODUCTION

In this experiment you will study the emission and absorption of *gamma rays* by an atomic nucleus: photons associated with nuclear quantum state transitions. Because a high energy gamma ray photon also has a large momentum, its absorption or emission results in a significant amount of recoil by the nucleus. The kinetic energy required to generate the recoil of a single nucleus would reduce the energy of the emitted photon so much that it would have insufficient energy to be absorbed by another, identical nucleus. How then can we actually observe such a process? The Nobel Prize winning efforts of Rudolph Mössbauer demonstrated a way to do so by taking advantage of the quantum-mechanical nature of the vibration states of atoms in a crystal.[1]

You will investigate this phenomenon using a 14.4 keV transition[2] between nuclear states of a rather uncommon but stable isotope of iron: ^{57}Fe (57 total *nucleons*,[3] whereas the most abundant iron isotope is ^{56}Fe). This isotope is the product of the decay of a radioactive cobalt isotope, ^{57}Co. By employing the Mössbauer effect you can use an absorber containing ^{57}Fe as a probe to generate a very precise energy spectrum of this 14.4 keV transition, revealing a resonance with an extremely high quality factor, Q.

Nuclear transitions and gamma rays

Recall that atomic electrons occupy different wave functions (levels and orbitals) in the Coulomb field of the atom. The lowest energy configuration of the atomic electrons is the atom's *ground state*; other configurations result in excited atomic states which eventually decay to the ground state, usually by the emission of one or more photons. Energy levels of the outer electrons (the "valence" electrons) are separated by energies of a few eV (give or take an order of magnitude), so transitions among these electrons involve photons from the near infrared to the ultraviolet (~1000 nm to 100 nm wavelengths). The inner shell electrons of all but the lightest atoms are much more strongly bound, however, resulting in transition energies of 10^3–10^5 eV (1 to 100's of keV). Photons in this energy range are called *x-rays*.

An atom's nucleus is a bound state of several nucleons — protons and neutrons — each of which has a rest energy of about 0.93 GeV; an electron's rest energy is 511 keV, over 1800 times

---

[1] The German physicist Rudolph Mössbauer won the 1961 Nobel Prize for his discovery of what eventually came to be called the *Mössbauer effect*. We'll have more to say about him later.

[2] Energies will be expressed in *electron volts* (eV), with keV = 10^3 eV, GeV = 10^9 eV, etc. A particle's mass will be stated as its *rest energy*: mc^2, where m is the particle's *rest mass*. Although in the text we may talk of a particle's mass, we will usually mean its rest energy (in eV).

[3] The protons and neutrons making up an atomic nucleus are collectively referred to as *nucleons*. The total number of nucleons, W, determines the atomic weight, whereas the number of protons, Z, determines the nuclear charge and the element it represents. ^{57}Fe has 26 protons (thus making it an isotope of iron) and 31 neutrons. Natural iron contains about 2% ^{57}Fe.

smaller. Nuclear structure is analogous to atomic electron structure: nucleons are arranged in various levels to determine a nuclear state. Excited nuclear states can decay by the emission of photons (among other processes). Because an atomic nucleus is so much smaller than an atom ($\sim 10^{-5}$ Å vs. ~1 Å), the transitions involve much higher energies (10's to 1000's of keV, many orders of magnitude higher than those associated with visible light). As mentioned earlier, the photons associated with nuclear transitions are called gamma rays.[4]

The internal nuclear state transitions described above involve electromagnetic interactions among the nucleons and therefore can include the emission or absorption of gamma ray photons. Other interactions (e.g., *forces*) are also important to the dynamical behavior of the nucleus: the *strong force*, which binds the nucleons together (and even makes possible the formation of individual nucleons) and the *weak force*, which causes radioactive *beta decay*. In our case, a ^{57}Co nucleus will eventually fall to a lower energy state by using the weak force to become ^{57}Fe. Following this transformation, it can lose additional energy through electromagnetic transitions until it finally reaches a stable state, the *ground state* of ^{57}Fe. This transformation of ^{57}Co into the ground state of ^{57}Fe involves the energy states (also called *levels*) and transitions diagrammed in Figure 1.

The weak-force mediated transition from ^{57}Co to ^{57}Fe is indicated by the diagonal arrow in Figure 1. This process is an example of *inverse beta decay*, wherein a nuclear proton interacts with one of the atom's inner-shell electrons (this interaction is also commonly called *electron capture*, *EC*). The electron's charge is transferred to the proton, changing the proton into a neutron. The electron is simultaneously converted into an electron *neutrino*, which then escapes the nucleus (and very likely the solar system as well!) with energy equal to the difference between the rest energies of the parent ^{57}Co atom and its ^{57}Fe daughter.

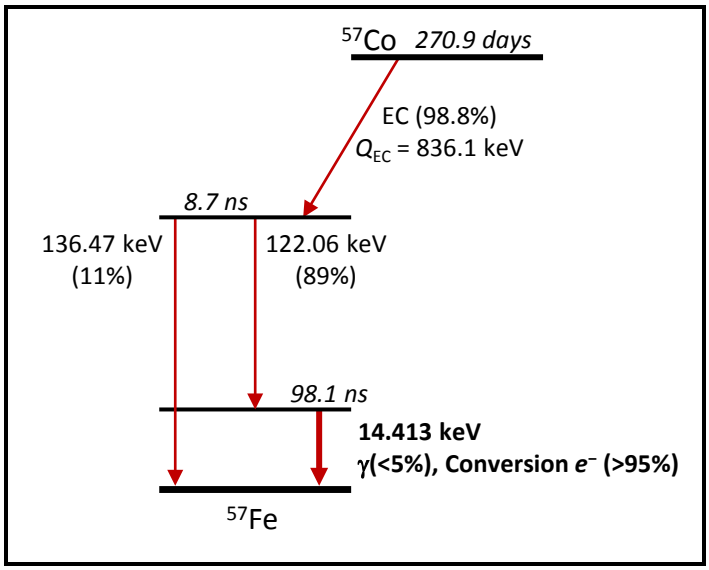


Figure 1: ^{57}Co decay scheme showing the relevant ^{57}Fe levels and emissions. The 14.413 keV transition is used for this experiment. State *half-life* times are also listed.

Following electron capture the resulting ^{57}Fe nucleus reaches its ground state via the possible electromagnetic transitions shown by the vertical arrows in Figure 1. Each of these transitions is accompanied by a gamma ray photon emission, except for the one we're most interested in: that

[4] Named by the British physicist Ernest Rutherford, gamma radiation was discovered by Paul Villard in 1900 in Paris. Rutherford with his colleagues Hans Geiger and Ernest Marsden discovered the atomic nucleus in 1911.

from the 14.4 keV excited state to the ground state. Over 95% of the time this transition takes place by a process called *internal conversion*: excess nuclear energy is transferred to an atomic electron through the Coulomb force between them, ejecting the resulting *conversion electron* from the atom. Only about 5% of the time is the 14.4 keV transition accomplished by photon emission.

Whenever an inner-shell atomic electron is lost, either through electron capture or internal conversion, the atom's remaining electrons cascade downward to fill the void, in the process emitting x-rays and other photons. For our purposes, the most important of these atomic photon emissions is the 6 keV x-ray created by an electron transition into an iron atom's inner shell. In addition, the 122 or 136 keV gamma rays emitted during the ^{57}Co decays can ionize atoms in the surrounding material, ejecting inner electrons and subsequently generating additional x-rays. The detector we will use during this experiment can very efficiently respond to all of these x-ray emissions.

THEORY: LINE WIDTHS, RECOIL, AND THE MÖSSBAUER EFFECT

Excited state lifetime and line width

Any introduction to quantum mechanics will demonstrate that an isolated, bound system (such as a particle in a potential well) has one or more *stationary states*: states of well-defined system total energy which are *eigenstates* of the system's *Hamiltonian operator*. The state with lowest total energy is called the system's ground state, and the others are the system's various excited states. According to this most simple theory, an isolated system prepared in an eigenstate of the Hamiltonian will remain in that state indefinitely, and its time-dependent state vector $|\Psi(t)\rangle$ will be given by $|\Psi(t)\rangle = e^{-i\omega_0 t}|\Psi_0\rangle$, where $|\Psi_0\rangle$ is a time-independent state vector and $\hbar\omega_0 = E_0$ is the well-defined energy of the state (the eigenvalue of the Hamiltonian corresponding to the state $|\Psi_0\rangle$).[5] Thus according to this simple theory a system prepared in a state with energy greater than that of its ground state will never decay to a lower energy (if it were truly isolated, then of course there would be no way for it to rid itself of the excess energy).

Actually, it appears that no system can be perfectly isolated because, as a more sophisticated and complete theory will suggest, even empty space (the "vacuum") is not a benign actor with no effect on a quantum system.[6] It is observed that excited quantum states of a system eventually spontaneously decay to ever lower energy states until the system's ground state is reached, its

---

[5] The concept of the *state vector* was integral to the *matrix mechanics theory* of quantum phenomena, first conceived by the German physicist Werner Heisenberg and later formulated by him and his colleagues Max Born and E. Pascual Jordan in a series of seminal papers in 1925; Heisenberg was awarded the 1932 Nobel Prize "for the creation of quantum mechanics." The British physicist Paul Dirac introduced the modern *bra* and *ket* notations for quantum state vectors, as well as the *Dirac delta function* $\delta(\vec{r})$ and the notation $\hbar$ for $h/2\pi$. Dirac shared the 1933 Nobel Prize with Erwin Schrödinger, inventor of quantum *wave mechanics* and his famous wave equation.

[6] *Quantum field theory* provides an example of such a theory.

excess energy being lost through whatever channels exist connecting the system with the outside world.[7] In the case of simple, relatively "pure" quantum systems, the probability of a system remaining in any particular excited state falls exponentially with time, so that $\left|\langle\Psi_0|\Psi(t)\rangle\right|^2 \propto e^{-t/\tau}$, where τ is a constant called the state's *mean lifetime*. Therefore the actual time dependence of any particular excited state vector, otherwise undisturbed, is observed to be

$$|\Psi(t)\rangle = e^{-(\gamma + i\omega_0)t}|\Psi_0\rangle \tag{1}$$

where $\gamma = 1/(2\tau)$, and $\hbar\omega_0 = E_0$ becomes a nominal energy associated with the excited state. What this expression means is that the time that any particular system will spend in this excited state before decaying is random, but the probability that it will remain excited without decaying relaxes exponentially toward 0 as time goes on.

Taking the Fourier transform of (1) with respect to time and normalizing provides the excited state's complex state vector frequency spectrum:

$$|\Psi(\omega)\rangle = \frac{\sqrt{\gamma/\pi}}{\gamma + i(\omega - \omega_0)}|\Psi_0\rangle$$

Using $\gamma = 1/(2\tau)$, squaring, and scaling the frequency by $\hbar$ yields the state's *energy spectrum*:

$$|\Psi(E)|^2 = \langle\Psi(E)|\Psi(E)\rangle = \frac{2\tau/(\pi\hbar)}{1 + (2\tau/\hbar)^2(E - E_0)^2} \tag{2}$$

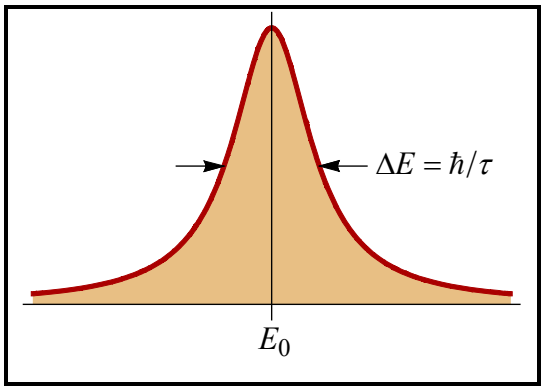


Figure 2: Lorentzian distribution of the energies of an excited state with nominal energy E_0 and mean lifetime τ. ΔE is the *full width at half maximum* (FWHM) of the peak.

This energy spectrum, or *distribution*, shown in Figure 2, is known by physicists as a *Lorentzian* and by mathematicians as a *Cauchy distribution*.[8] Equation (2) provides the *probability density function* (PDF), $p(E)$, of the state's energy distribution. It is symmetric about its median or nominal energy E_0, and its *full width at half maximum* (FWHM) $\Delta E = \hbar/\tau$. This relation immediately leads to the (saturated) energy-time uncertainty relation:

$$\Delta E\, \Delta t = \hbar \tag{3}$$

---

[7] Of course, outside energy can also enter through such channels, keeping the system from perpetually resting in its ground state. Such considerations are in the realm of thermal physics.

[8] After Dutch Nobel laureate Hendrik Lorentz and 19th century French mathematician and physicist Augustin-Louis Cauchy. This distribution was first studied by the famous 17th century mathematician Pierre de Fermat. Interestingly, the Lorentzian distribution has an undefined statistical mean and standard deviation, because the integrals defining these statistics do not converge.

where in this case $\Delta t = \tau$, the state's mean lifetime. The Lorentzian "line shape" is characteristic of carefully-measured spectral lines of atomic emissions and of a variety of other sorts of resonant responses. This *resonant emission process* provides a quantum mechanical analog of the time evolution of the energy stored in a classical, damped harmonic oscillator (its *transient response*).

The proper interpretation of equation (2) is that it describes the probability distribution of the measured energies of the excited state, so that a state with a finite lifetime no longer has a well-defined energy (unlike a true stationary state). This means that, for example, photons emitted during decays of such states will exhibit a distribution of energies given by (2). Thus a determination of the distribution of emitted photon energies measures the state's energy distribution. Conversely, equation (2) describes the relative probabilities that incoming photons of various energies will be absorbed and excite the system into that state.

Consider the combined events of photon emission as one system's excited state decays followed by absorption of the photon by another system, transitioning it into an excited state. If the emitter's excited state has an energy distribution described by the PDF $p_e(E, E_e)$, where E_e is the nominal energy of its excited state, then $dP = p_e(E, E_e)\,dE$ is the differential probability that the state will emit a photon within dE of energy E when it decays. Then again let $p_a(E, E_a)$ be the probability density that a properly aimed, emitted photon with energy E will excite an absorber with a nominal excited state energy within dE_a of E_a (and be absorbed). The overall probability density that an emission from the decay of a state with nominal energy E_e will be followed with an absorption by a state with nominal energy E_a is then proportional to the integral:

$$p_{e\to a}(E_e, E_a) = \int_{-\infty}^{\infty} p_e(E, E_e)\, p_a(E, E_a)\, d(E)$$

That this expression is a probability density may be confirmed by checking its units. The Lorentzian distribution (2) depends only on the squared difference $(E - E_0)^2$, so it is symmetric in E and E_0. Thus for the case of two Lorentzian distributions with FWHM line widths ΔE_e and ΔE_a the integral of the above expression is straightforward, and

$$p_{e\to a} \propto \frac{1}{(\Delta E_e + \Delta E_a)^2/4 + (E_e - E_a)^2} \tag{4}$$

Therefore the resulting *convolution integral* of two Lorentzian distributions is a Lorentzian in the difference of the two nominal excited state energies, with FWHM given by the sum of the two states' FWHM values, as shown in Figure 3 on page 6.

Emission and absorption recoil kinematics

The discussion of the previous section neglected to consider momentum conservation during photon emission and absorption. We now take up this issue. When a system decays by emitting a

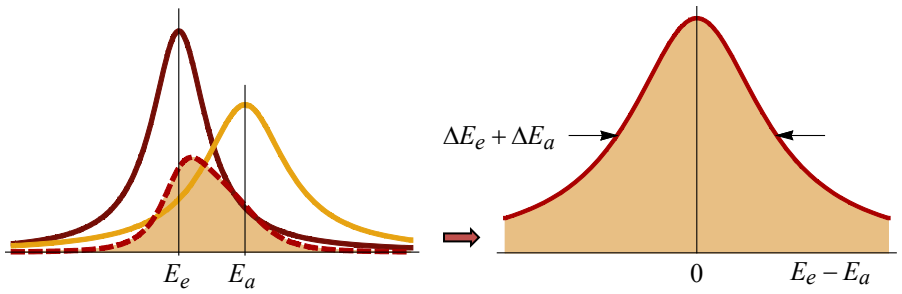


Figure 3: *Convolution* of emitter and absorber Lorentzian distributions. The left graph shows the two distributions for an assumed E_e and E_a. The shaded area is the integral of the product of the two Lorentzians. Plotting the value of this integral as a function of their energy difference results in the right graph, a Lorentzian with FWHM equal to the sum of the emitter and absorber widths.

photon, the photon not only carries away energy but also linear momentum.[9] A photon with energy $E_\gamma = \hbar\omega$ also carries linear momentum $p_\gamma = \hbar k = E_\gamma / c$. To conserve the total momentum of the emitter+photon system, the emitter must recoil with an equal and opposite momentum, requiring some energy from the emission process. Similarly, upon absorption the absorber must recoil with the photon's momentum, requiring some energy from the photon to support this center of mass motion.

Let's do the math.[10] A system has rest energy M and excited state energy $M^* = M + E_e$. It decays by emitting a photon with energy k. Consider the decay process first. The original, excited system is at rest with energy M^*, so its 4-momentum is $(M^*, 0)$. 4-momentum is conserved, so following the decay the emitter+photon will still have the same total 4-momentum. The photon's 4-momentum is $(k, \mathbf{k})$, therefore the emitter's is $(E_M, -\mathbf{k})$, with $E_M = M^* - k$. Now the basic relativistic relationship between the components of any object's 4-vector $(E, \mathbf{p})$ is that $E^2 = M^2 + p^2$, where M is the object's rest energy. Therefore:

$$(M^* - k)^2 = (E_M)^2 = M^2 + k^2 = (M^* - E_e)^2 + k^2$$

$$\therefore 2M^* k = 2M^* E_e - E_e^2$$

$$\boxed{k = E_e\left(1 - \frac{E_e}{2M^*}\right)} \qquad (5)$$

So, as expected, the emitted photon's energy is a little less than the decay energy E_e. Similarly, a system at rest with 4-momentum $(M, 0)$ absorbs a photon $(k, \mathbf{k})$ to enter the excited state

---

[9] The photon also carries away angular momentum, but that need not concern us here. Experiments 27 and 29 do take up this issue.

[10] The relativistic kinematics are discussed in more detail in Physics 7 General Appendix A: *Relativistic Kinematics*, http://www.sophphx.caltech.edu/Physics_7/General_Appendix_A.pdf. We follow the notation and conventions of that document: we use units wherein the speed of light $c \equiv 1$, rest masses are referred to by their corresponding rest energies, and a photon's energy and momentum both use the symbol k. The 4-momentum of a particle at rest is then (energy, momentum) = $(M, 0)$; for a photon: $(k, \mathbf{k})$, with bold font for a spatial 3-vector (this differs from Appendix A, which uses an arrow over a 3-vector).

$M^* = M + E_a$. Following the absorption, the system's state is (E_{M^*}, **k**), with $E_{M^*} = M + k$. Following a calculation similar to the above (see the Prelab Problems), the relationship between k and E_a is:

$$\boxed{k = E_a\left(1+\frac{E_a}{2M}\right)} \tag{6}$$

So to support the absorbing system's recoil, the incoming photon must have a little more energy that the transition energy E_a.

Just how important is this effect? First consider an atomic emission and absorption, for example the neon 626.6 nm normal Zeeman line used in Experiment 27. Neon has a rest energy of $20.0 \times 0.93 = 18.6\,\text{GeV}$. The transition energy is $hc/(626.6\,\text{nm}) = 2.0\,\text{eV}$. Therefore the recoil corrections given in equations (5) and (6) are $\sim 10^{-10}$ eV. The lifetime of the excited state is about $\tau \sim 10^{-8} - 10^{-7}$sec, so from equation (3) the line width $\Delta E \sim 10^{-8} - 10^{-7}$eV, hundreds to thousands of times greater than the recoil energy. This means that the recoil effect of a typical visible light photon emission is completely negligible.

What about the ^{57}Fe 14.4 keV nuclear transition? With a mean lifetime of approximately 150 nsec, the line width is again $\Delta E \sim 10^{-8}$ eV. The recoil energy requirement changes the photon energy by $(14.4\,\text{keV})^2/(2\times 53.6\,\text{GeV}) = 2\times 10^{-3}\,\text{eV}$, 10^5 times greater than the line width! The photon leaves the decay with this much less energy than the transition energy, but it requires this much more energy to excite a transition in another nucleus. Thus the recoil energy losses would seem to make emission followed by subsequent absorption of this photon very nearly impossible. Luckily, the Mössbauer effect will come to the rescue!

The Mössbauer effect

An atom of mass M in a solid is part of a crystal and is bound to the atoms surrounding it. To lowest order, we can treat the atom as sitting in a quantum harmonic oscillator potential well established by this binding to its neighbors. In its ground state, such an oscillator has a wave function which is Gaussian in both position and momentum space; we are concerned with its behavior in momentum space. The atom's ground state momentum space wave function goes as

$$\begin{aligned}\psi_0(\mathbf{p}) &\propto \exp\left(\frac{-p^2}{2M\hbar\omega_0}\right) = \exp\left(\frac{-p_x{}^2}{2M\hbar\omega_0}\right)\exp\left(\frac{-p_y{}^2}{2M\hbar\omega_0}\right)\exp\left(\frac{-p_z{}^2}{2M\hbar\omega_0}\right) \\ &= \psi_0(p_x)\,\psi_0(p_y)\,\psi_0(p_z)\end{aligned} \tag{7}$$

The angular frequency ω_0 is the resonant frequency of the harmonic oscillator, and $\hbar\omega_0$ is the spacing between its energy levels.[11] Since this wave function factors into a product of Gaussian

[11] Actually, there are many resonant frequencies corresponding to the many vibration modes of the crystal. We consider only the highest frequency mode here, roughly corresponding to each atom vibrating out of phase with its

wave functions for each of the three Cartesian coordinates, we need only consider that component parallel to the direction of the recoil momentum. Now consider the effect of the impulse of momentum generated by the recoil following a photon emission or absorption. Immediately following the recoil, the atom's momentum space wave function is displaced by the recoil momentum k as illustrated in Figure 4.[12]

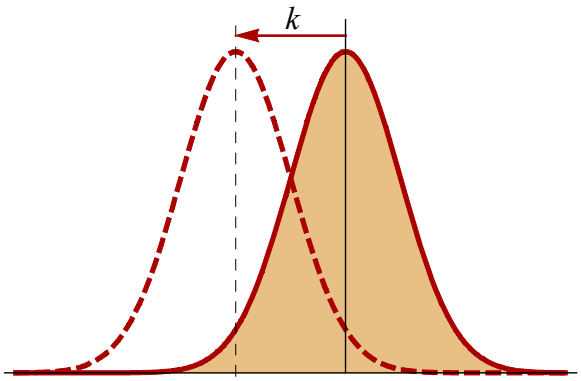


Figure 4: The momentum space ground state wave function of an atom in a potential well is suddenly displaced by the momentum impulse *k* generated by gamma ray photon emission or absorption. The displaced wave function (dashed profile) is not an energy eigenstate of the original harmonic oscillator potential of the atom in the stationary crystal.

Of course, the entire crystal will absorb this extra momentum k, acquiring the absolutely microscopic center-of-mass kinetic energy $k^2/(2M_{\text{crystal}})$, on the order of 10^{-20} of the recoil energy required by a single atom of mass M. On the other hand, the sudden recoil of the atom may excite a higher harmonic oscillator vibration state of the crystal, requiring additional energy from the photon absorption or emission process (eventually turned into heat in the crystal). $|\Psi_k\rangle$, the displaced wave function of the single recoiling atom shown in Figure 4, can be expressed as a coherent superposition of the vibrational energy eigenstate wave functions of the recoiling atom. The square of the matrix element $|\langle\Psi_k|\Psi_n\rangle|^2$ then gives the probability that the recoiling atom is excited into its nth vibration state, requiring an additional energy of $n\hbar\omega_0$. But the probability that the atom *is not* excited from its ground vibration state by the recoil is given by $|\langle\Psi_k|\Psi_0\rangle|^2$, where $|\Psi_0\rangle$ is the atom's original, ground vibration state shown by the shaded wave function in Figure 4. This matrix element is then:

$$\langle\Psi_k|\Psi_0\rangle = \sum_p \langle\Psi_k|p\rangle\langle p|\Psi_0\rangle = \int \psi_0^*(p+k)\,\psi_0(p)\,dp = \exp\left(\frac{-k^2}{4M\hbar\omega_0}\right)\cancel{\langle\Psi_0|\Psi_0\rangle}^{(=1)} \qquad (8)$$

---

neighbors. To lowest order, each crystal vibration mode acts as an independent quantum harmonic oscillator. If the α^{th} vibration mode with resonant frequency ω_α is in its nth excited state (with $n = 0$ corresponding to its ground state), then we say that the crystal contains n *phonons* of frequency ω_α. In general, thermal excitations and other sources of energy or vibration will excite many modes. In our gross simplification, we assume only a single vibration mode originally in its ground state.

[12] Another assumption here: we assume that the impulse generated by the gamma ray absorption or emission is short compared to the vibration time scale ω_0^{-1}. This, as it turns out, is actually an excellent assumption.

Let's plug in some numbers: according to equations (5) or (6), the $k^2/(4M)$ in equation (8) is half of a single, isolated ^{57}Fe atom's recoil energy, which we previously calculated to be $2\times10^{-3}\,\text{eV}$ for the 14.4 keV nuclear transition. Inserting this value along with $\hbar\omega_0 \sim 10^{-2}\,\text{eV}$ (see the Prelab Problems) and squaring the matrix element, the probability $\left|\langle\Psi_k|\Psi_0\rangle\right|^2$ is ~80%! Of course this has to happen for both the emission and for the subsequent absorption, so the joint probability would drop to (80%)^2, or ~60%. With all of the approximations and simplifications that went into the argument leading up to this calculation, that estimate is certainly not very accurate. Nevertheless, it is clear that $\left|\langle\Psi_k|\Psi_0\rangle\right|^2$ represents a significant probability.[13]

When the above scenario is realized, an entire crystal of 10^{20} or more atoms recoils as a unit, with no extra energy required for internal vibrations. The resulting recoil mass to be used in equations (5) or (6) is thus M_{crystal}, and the recoil energy required becomes microscopically smaller than the $10^{-8}\,\text{eV}$ line width of the 14.4 keV nuclear transition. This is the phenomenon now called the *Mössbauer effect*. As an added bonus the random thermal motion of the emitter is effectively "frozen out" for a large fraction of the gamma ray emission and absorption events. Because the probability of this behavior is quite significant, we can make use of it to perform the experiment of 14.4 keV nuclear photon emission followed by subsequent nuclear absorption.

The discovery by Rudolph Mössbauer in 1957 of what he called *recoilless nuclear resonant emission and absorption* provided this major breakthrough in the detailed study of certain nuclei. Mössbauer was a graduate student working in Heidelburg, Germany, at the time of his discovery; his PhD thesis of 1958 described his work. Harvard's Robert Pound and Glen Rebka quickly seized on Mössbauer's technique to perform an experiment in 1959–1960 to measure the gravitationally-induced frequency shift of gamma ray photons falling through a vertical distance of only about 21 meters; their astonishing result provided the first accurate experimental verification of Einstein's *general theory of relativity* (interestingly, Pound, also a co-discoverer at Harvard of *nuclear magnetic resonance*, was a tenured professor who had never earned an advanced degree).

Mössbauer came to Caltech in 1960 and became a professor in 1962 (he returned to his alma mater in Germany in 1964). In 1961 he was awarded the Nobel Prize in physics for his 1957 discovery (probably because of the profound importance for physics of Pound and Rebka's experiment). His discovery subsequently led to the thriving field of *Mössbauer spectroscopy*, a highly-sensitive, analytic probe of molecular and crystalline structures used by researchers in fields as diverse as molecular biology and geophysics.

---

[13] More realistically, at the time of a nuclear emission or absorption the crystal will contain many phonons occupying many different crystal vibration modes with different frequencies and excitation energies. Mössbauer's recoilless nuclear emission and absorption would occur only if the crystal's original phonon content is unchanged by the nuclear recoil, so that no energy is transferred into or out of any of the crystal vibration modes by the gamma ray emission or absorption. To read it straight from *der mund des pferdes*, you may want to look at Mössbauer's and D. H. Sharp's paper: https://doi.org/10.1103/RevModPhys.36.410, also available on the Caltech site https://authors.library.caltech.edu/11074/1/MOSrmp64a.pdf.

USING THE DOPPLER EFFECT TO MAP A SPECTRAL LINE

Design of the experiment

The ^{57}Co decay to ^{57}Fe provides a source of 14.4 keV photons as the daughter nuclei decay to their ground states. By taking advantage of the Mössbauer effect, these emitted photons should have a Lorentzian distribution of energies with FWHM $\Delta E_e \approx 10^{-8}$ eV around the nominal energy $E_e \approx 14.4$ keV, as discussed previously. To map out the Lorentzian shape of this 14.4 keV nuclear transition, we need an extremely precise way to measure the energy distribution of the emitted photons. Looking again at Figure 3 on page 6, we see that if we had an absorber whose nominal energy E_a could be precisely adjusted around 14.4 keV, then the rate that it absorbs the emitted photons could be used to map out the convolution of the emitter and absorber distributions as a function of $E_e - E_a$.

This experiment is designed to do just that by using the Doppler effect to vary E_a (or, equivalently, E_e). Figure 5 illustrates the idea. The detector has a broad energy range over which it can detect photons, so any photons emitted by the source which enter the detector will be detected with equal efficiency. Interposing the absorber will cause some fraction of the photons to be absorbed, reducing the rate of photon arrival at the detector. As the effective E_a of the absorber is varied by changing its velocity v toward or away from the source, so will its photon absorption fraction. Increased absorption corresponds to a lower photon rate at the detector, so the convolved Lorentzian distribution as a function of $E_e - E_a$ takes the form of a decrease in detection rate as shown in the plot in Figure 5, creating an *absorption line* profile.

Doppler shift of photon energies

A shift in frequency of a photon is equivalent to shifting its energy, because $E = \hbar\omega$, as we used to derive equation (2). To map out a Lorentzian distribution requires a relative shift in frequency of a few times $\Delta\omega/\omega = \Delta E/E \approx 10^{-8}\,\text{eV}/10^{4}\,\text{eV} = 10^{-12}$. This tiny fractional change in frequency

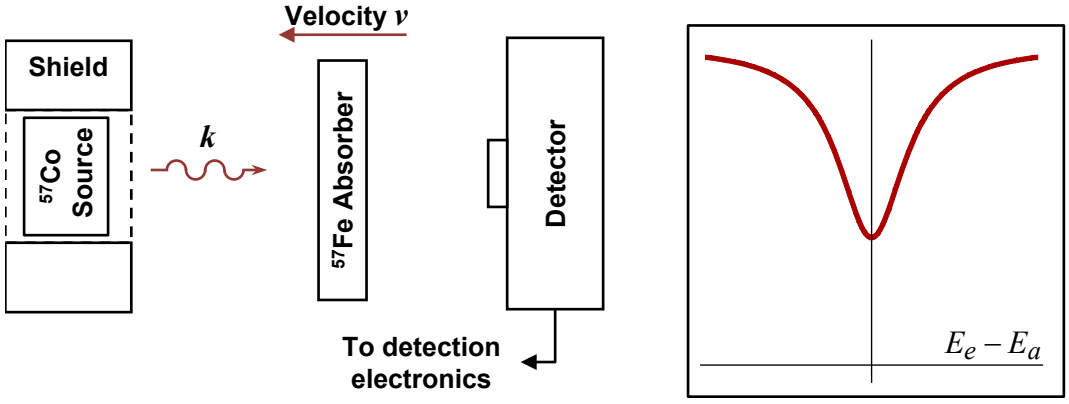


Figure 5: Conceptual setup of the experiment. By changing the relative velocity v between the absorber and the source, emitted photons at energy k are Doppler shifted to higher or lower energy as seen by the absorber. This effectively tunes the energy difference $E_e - E_a$, changing the absorption rate and thus the rate that photons reach the detector (graph at right).

implies that only a small relative velocity v between source and absorber will be required to generate the Doppler shift in frequency. Therefore we do not need the relativistic Doppler formulas of Physics 7 General Appendix A, and we can use a simple, Newtonian analysis.

Looking at Figure 5, light with angular frequency ω travels rightward with phase velocity c, while the absorber travels leftward with velocity $|v| \ll c$. ($v > 0$ is toward the source, $v < 0$ is in the opposite direction). The wavelength of the light is $\lambda = 2\pi c/\omega$, and the relative velocity of the light wave and the absorber (from the point of view of the experimenter, at rest in the lab frame) is $c + v$. Therefore, the experimenter will see cycles of the wave pass the absorber at the rate $(c+v)/\lambda$, corresponding to an angular frequency $\omega' = 2\pi(c+v)/\lambda$. The relative motion has changed the light's frequency at the absorber: $\omega' = \omega(c+v)/c = \omega(1+v/c)$. This is, of course, the Doppler shift. The photon's energy would be shifted by the same fraction,

$$\frac{v}{c} = \frac{\Delta E}{E} \tag{9}$$

With a nominal energy of 14.413 keV,

Energy–velocity relation: $$\boxed{\frac{v}{\Delta E} = 0.2080 \, \frac{\text{mm/sec}}{10^{-8}\text{eV}}} \tag{10}$$

Doppler shifts from quite small velocities (fractions of a millimeter/sec) are required to map out the expected Lorentzian energy distribution of the 14.413 keV nuclear transition in ^{57}Fe.

Isomeric shift

The source you will use contains ^{57}Co atoms embedded in a foil of either rhodium or palladium metal (depending on the manufacturer of the ^{57}Co radioactive source). The absorber will be a thin foil of nonmagnetic stainless steel (^{57}Fe makes up 2.12% of natural iron). The chemical bonding of a ^{57}Co or a ^{57}Fe atom to its neighbors in a solid introduces distortions in its various atomic electron wave functions. These distortions cause the electromagnetic field surrounding the nucleus to be slightly altered, which in turn very slightly shifts the differences between the energy levels of the nucleon states. These shifts differ for ^{57}Fe nuclei embedded in different materials, so a slight Doppler shift offset is required to adjust the nominal emitted gamma energy to that of the absorber. This small *isomeric energy shift* ΔE_{iso} between source and absorber nominal energies requires a small relative velocity offset v_{iso} to provide a compensating Doppler shift in the gamma ray energies.

By convention, ^{57}Fe isomeric energy shifts are defined relative to the actual energy of the 14.413 keV nuclear transition of ^{57}Fe when embedded in a crystal of pure, natural iron at room temperature (called *α-iron*, with a *body-centered cubic* crystal structure). The isomeric shifts of the ^{57}Fe transition have been measured for many materials; selected results are shown in Figure 6 on page 12.

Isomeric shifts

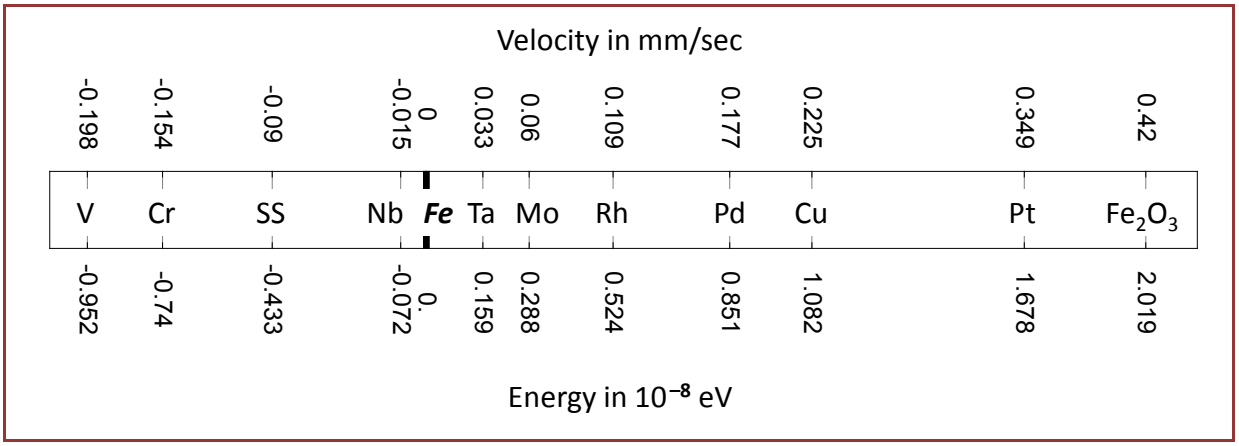


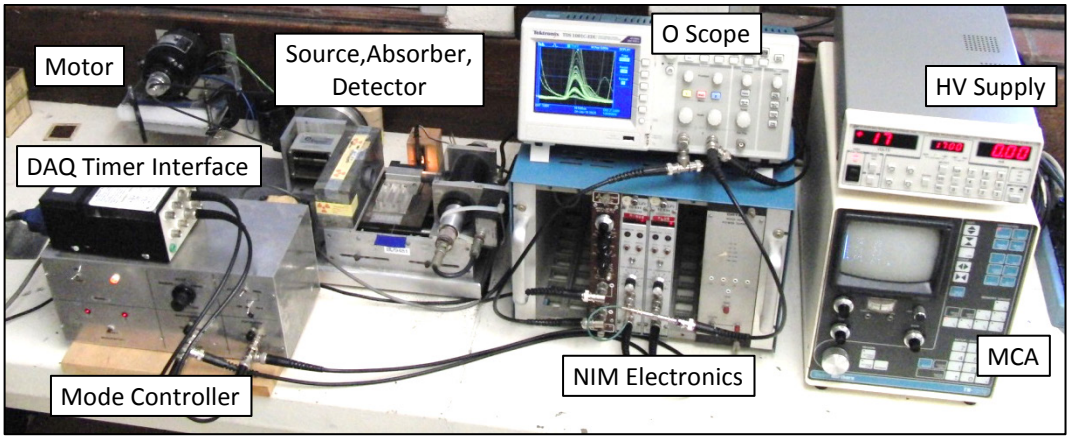
Figure 6: A selection of isomeric shifts of the 14.413 keV nuclear transition when ^{57}Fe or ^{57}Co is alloyed with various materials. The shifts are with respect to the energy of the 14.413 keV transition in α-Iron (*Fe* in the chart). "SS" denotes type 302 stainless steel. Uncertainties are: ±0.02mm/s for SS; ±0.002mm/s for Cu, Pd, Rh; ±0.006mm/s for Pt; ±0.009mm/s for Cr; ±0.01mm/s for the rest.

The above chart shows the relative energies and corresponding Doppler velocities of the 14.4 keV excited state when ^{57}Fe is embedded in the various materials. If the absorber has a more negative energy than the emitter, then clearly a "red shift" of the emitted photons will be necessary to maximize absorption, the difference in the chart velocities telling you the magnitude of the shift. Blue shift of the photons' energies is required if the absorber has a higher energy than the emitter.

EXPERIMENTAL APPARATUS AND SOFTWARE

Hardware

The following figures provide brief descriptions of the system components and their controls.



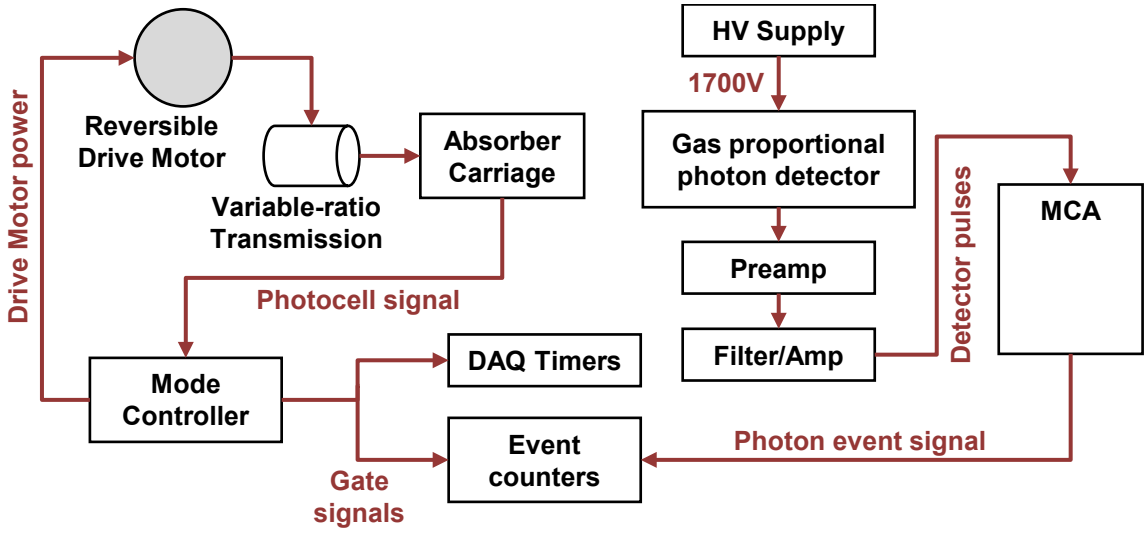


Figure 7: General arrangement and system block diagram of the apparatus. The source, absorber and detector assembly is the heart of the system (Figure 8), the mode controller is its brain (Figure 9). The drive motor is activated by the mode controller and moves the carriage holding the absorber so that a Doppler shift is generated. The photon detector is powered by a high-voltage supply and outputs pulses proportional to the detected photon energies. The amplified pulses are sorted by the MCA (Figure 11). When properly set the MCA generates an output pulse whenever a 14.4 keV photon is detected. The mode controller also generates a gate signal to activate the appropriate DAQ timer and NIM event counter (Figure 10) as the absorber moves at a constant velocity toward or away from the cobalt source. Count data is collected as the absorber moves through a distance of approximately 25 millimeters. The photocell signal from the carriage assembly (Figure 8) is used by the mode controller to generate the data acquisition gate signal.

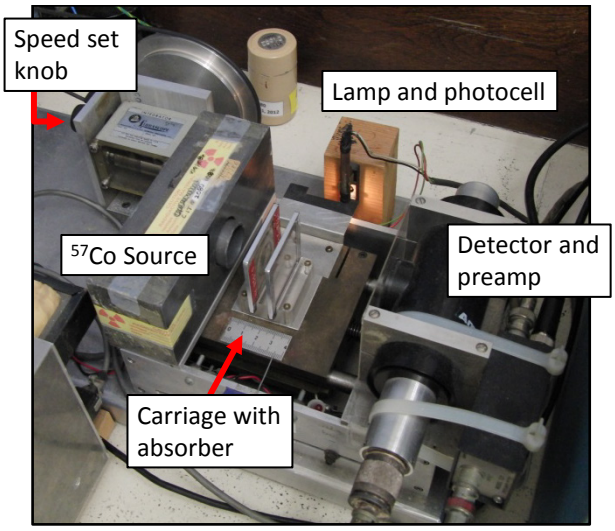


Figure 8 (right): Detail showing the heart of the apparatus. The speed knob controls the variable-ratio transmission, adjusting the rotation rate of the drive screw which moves the carriage containing the absorber. The gas proportional detector is connected to a preamp which supplies it with power and generates an output voltage pulse following each photon detection event. The lamp and photocell arrangement determines the distance over which the carriage travels while acquiring data. It also ensures that the carriage speed has stabilized before data acquisition begins.

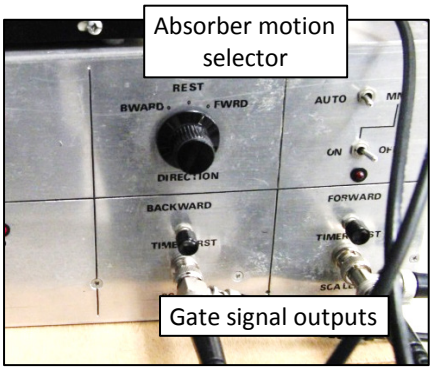


Figure 9 (left): The Mode Controller has a rotary switch which selects the direction of absorber motion: Forward (blue-shift), Backward (red-shift), or Rest (stopped). The controller then activates the motor in the proper direction. When the carriage photocell is activated, the controller sends a gate signal to activate the appropriate event counter and DAQ timer. The controller automatically stops the motor when the carriage motion limit is reached.

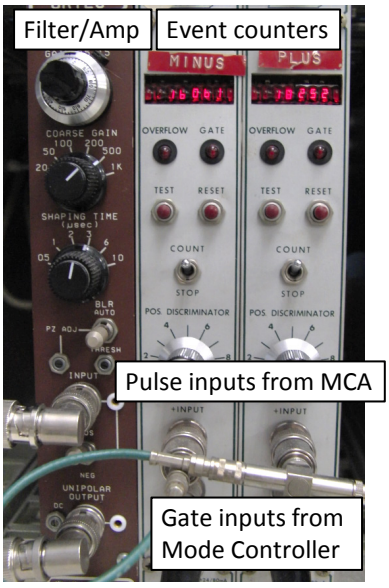


Figure 10 (right): The NIM (Nuclear Instrumentation Module) electronics should already be properly set. There are two event counters, one for blue-shifted absorber motion ("PLUS") and the other for red-shifted motions ("MINUS"). Event pulses are generated by the MCA (Figure 11), the counters are activated by the gate inputs from the Mode Controller. Reset the counts to zero using the individual Reset buttons on the counters.

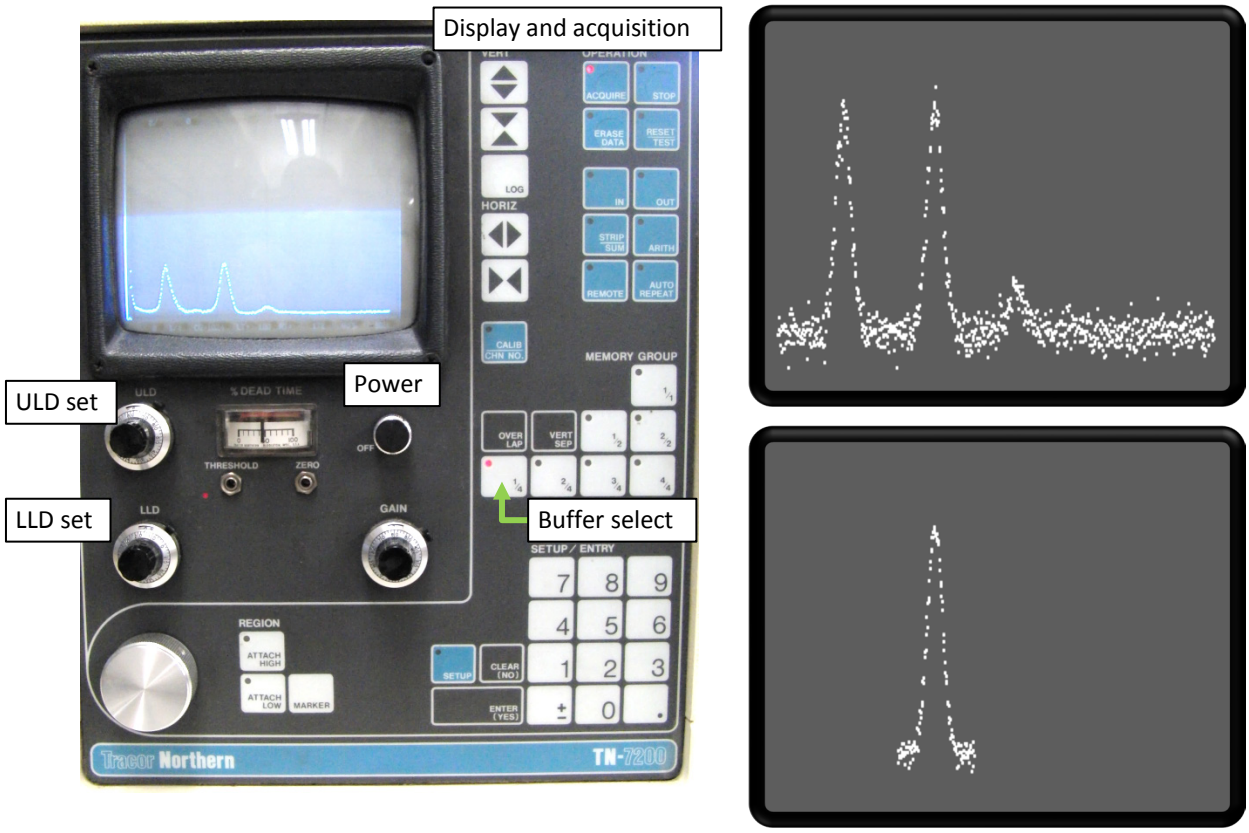


Figure 11: The MCA (left) must be powered on and a memory buffer selected (1/4 is a good choice). Use the display and acquisition controls to acquire photon energy spectra from the source.

The ULD (upper level) and LLD (lower level) discriminators must be set to isolate the 14.4 keV photon detections (right). The images are of simulated MCA spectrum displays. The amplified and filtered output from the gas proportional detector (Figure 8 and Figure 11) is a pulse with a peak voltage proportional to the energy of the detected photon. The MCA generates a histogram of pulse voltage (x-axis) vs. number of pulses seen (y-axis), producing a spectrum of the photon energies detected.

Upper image: the discriminator limits are wide, showing the entire spectrum from the source. The peaks are from the 6 keV and 14.4 keV emissions from iron and from the embedding matrix (Rh or Pd) x-rays at approximately 22 keV. The low-level background continuum counts are from the 122 and 136 keV gamma photons.

Lower image: ULD and LLD set to isolate the 14.4 keV peak. The MCA generates a digital pulse for each event detected within the LLD-ULD window. These digital pulses are routed to the NIM event counters so that count rates may be determined.

Software

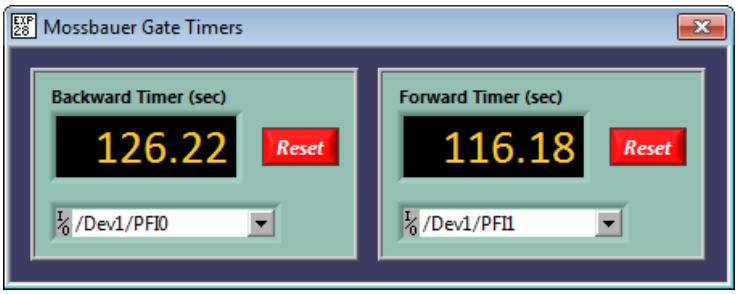
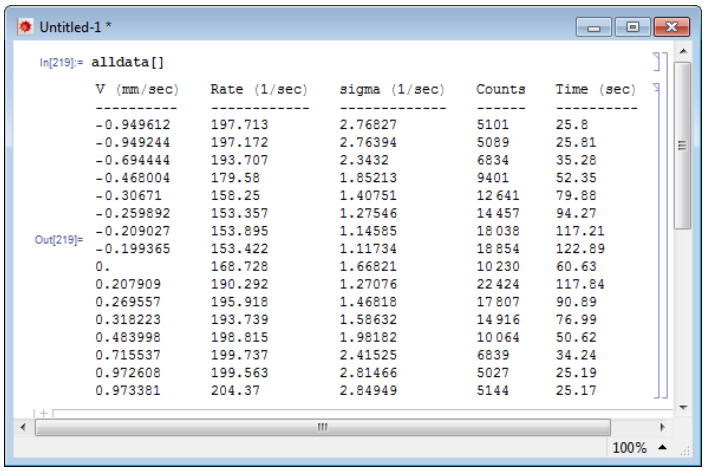


Figure 12: The Mossbauer Gate Timers application uses the mode controller gate signals connected to the computer DAQ to time the acquisitions. The times are then used to calculate the photon detection rates and the absorber velocities.



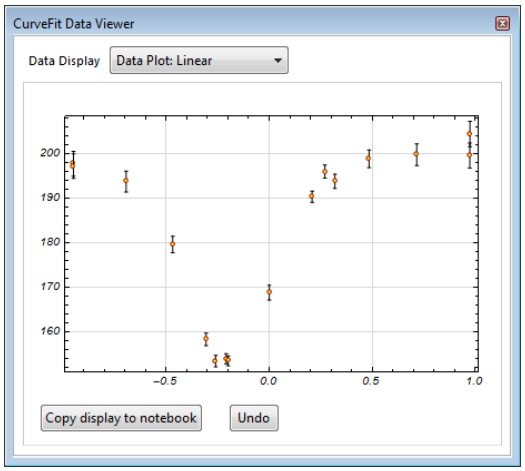


Figure 13: The *Mossbauer Helper* notebook for Mathematica® interfaces with *CurveFit* to help the experimenter record and process event counts and time measurements. The left image shows a notebook-generated table of processed (counts, time) measurements. The notebook automatically calculates absorber velocity, count rate, and count rate uncertainty (assuming Poisson count statistics) from raw data. The results are then automatically merged into the current *CurveFit* data set, shown in the accompanying data plot. The x-axis is in mm/sec, the y-axis in counts/sec.

CAUTION

Do not stick your fingers into the opening in the lead brick containing the radioactive ^{57}Co source! Do not lean in close to the source to attempt to look at it.

You will handle lead shielding. Wash your hands when you leave the lab.

LAB PROCEDURE: SETUP AND INITIALIZATION

For each step of the procedure, first completely read the instructions and locate the relevant components and controls before attempting to perform the step.

Basic equipment and software setup

1. Remove any temporary lead shielding from in front of the ^{57}Co source and set it aside. Remove the stainless steel absorber and x-ray filter from the carriage assembly (Figure 8). Set the Mode Controller's motion control knob to **REST** and turn on the power to the Mode Controller (Figure 7 and Figure 9). The photocell lamp should illuminate (Figure 8). Check that the Mode Controller **AUTO**/**MNL** switch is in **AUTO**.
2. Check that the power to the NIM electronics is ON (switch is on the right panel of the NIM crate assembly, Figure 7). Turn on the oscilloscope. Turn on the MCA using the intensity knob just below the right edge of the MCA display screen (Figure 11). Select the **¼** buffer button and adjust the MCA display brightness. Press the MCA display and acquisition controls' **ERASE DATA** button to clear the MCA display (Figure 11). Unlock the **ULD** and **LLD** adjust controls and turn the **ULD** up by several turns and the **LLD** down to zero.
3. Check that the HV power supply is powered on and set to 1700V. Apply power to the photon detector using the switch at the left-bottom corner of the HV power supply panel (Figure 7). Pulses should appear on the oscilloscope display (Figure 7).
4. Start the *Mossbauer Gate Timers* application (Figure 12) and launch the *Mossbauer Helper* notebook. Execute the notebook's initialization cells. The notebook should then load and launch *CurveFit*. Arrange the windows on the computer display so that the gate timers window is visible along with the Mathematica notebook windows.

Carriage travel distance calibration

1. Remove the brown rubber drive belt connecting the variable-ratio transmission output to the carriage drive pulley so that you can turn the carriage drive pulley with your fingers.
2. By rotating the carriage drive pulley you can move the absorber carriage over its range of travel toward or away from the ^{57}Co source (Figure 8). A red LED illuminates whenever the photocell is illuminated, triggering data acquisition.
3. Use the distance scale on the carriage to determine the distance over which the LED is illuminated. Determine this distance as accurately as possible so that you may obtain accurate isomeric shift and absorption FWHM measurements.
4. Reconnect the rubber drive belt between the transmission and carriage pulleys.

Enter the measured distance (in millimeters) into your Mathematica data notebook using the *Mossbauer Helper* **`SetDist[ ]`** function.

MCA ULD and LLD adjustments

1. Remove the stainless steel absorber and x-ray filter (clear plastic) from the carriage (if still installed).
2. Acquire a photon spectrum using the MCA **AQUIRE**, **STOP**, and **ERASE DATA** buttons (Figure 11). Use the vertical display buttons just to the right of the display screen to adjust the displayed spectrum. Turn the **LLD** up from 0 a bit to remove any strong noise counts at the far left edge of the spectrum. The **% DEAD TIME** meter to the right of the **ULD** control should then read near 0%. You should obtain a spectrum similar to that in the upper-right image in Figure 11. If not, adjust the **ULD** and **LLD** controls to change the spectrum limits.
3. Identify the 6 keV and 14.4 keV photon peaks. Install the x-ray filter on the absorber carriage and acquire a new spectrum. The 6 keV peak's relative strength should be significantly smaller with the filter installed. Remove the filter once you have positively identified these peaks.
4. Acquire several spectra as you adjust the **ULD** and **LLD** controls until the MCA acquires only the 14.4 keV photon peak as shown in the lower-right image in Figure 11. Include a small range of energies on either side of the peak (as shown), so that if the overall gain of the pulse electronics should vary slightly, the 14.4 keV peak will remain in the window defined by the **ULD** and **LLD** control settings.

Lock the **ULD** and **LLD** controls so that they can't be accidentally changed.

Final setup adjustments

1. Check that the motor drive belt is installed on the smallest motor pulley (the one at the very end of the motor drive shaft) and that the belt is on the largest pulley of the variable transmission. Check the alignment of the belt and pulleys so that the belt will not come off when the motor activates. Adjust the position of the motor on its mounting platform to align the belt and pulleys. The drive belt should not be so tight that it stretches the belt material.
2. Install the stainless steel absorber and the x-ray filter on the carriage assembly. The absorber should be closer to the radioactive source, the x-ray filter closer to the detector. Ensure that they fit snugly into their mounting slots.
3. Turn the speed set knob all the way counter-clockwise, then add a 1/2–turn clockwise from its limit. Use the Mode Controller motion control knob to start the drive motor. Check that the belts are properly installed and aligned. Now adjust the speed set knob until the transmission output shaft is not rotating and that the absorber carriage has stopped moving. Return the Mode Controller motion control knob to **REST**.

If the motor drive belt is installed on the proper pulleys, then one full turn of the speed control knob will change the carriage speed by approximately 0.2 mm/sec: clockwise to increase the speed, counter-clockwise to reduce it.

4. Turn the speed control knob one full turn clockwise to change the carriage speed from 0 (set in step 3) to about 0.2 mm/sec. Select either **BWARD** or **FWRD** on the Mode Controller motion control knob and allow the absorber carriage to move to the appropriate limit of its range. The drive motor should automatically stop when that limit is reached.

Setup and initialization of the apparatus should now be complete.

LAB PROCEDURE: TAKING DATA

You will take data points by adjusting the speed set knob, then running the absorber both backward and forward at the selected speed. You will enter counts and time into a Mathematica notebook using the appropriate functions defined by the *Mossbauer Helper* notebook. The resulting *CurveFit* data plot will help you decide what speeds you need to fill out your spectrum of the Mössbauer absorption feature.

Let the absorber run through its complete range and wait for the drive motor to turn off before starting the absorber back in the opposite direction.

Start with the absorber speed set to approximately 0.2 mm/sec, as you set it at the end of the setup procedure. Reset the two NIM counters and the two timers of the *Mossbauer Gate Timers* application. When you select either **BWARD** or **FWRD** to start the absorber motion, the motor will start. The corresponding gate timer and event counter will then start when the carriage photocell

is activated. The counting and timing will stop, and after a few more seconds the motor will turn off. Don't start the absorber motion in the other direction until the motor has completely stopped.

Important tip: enter each data point into Mathematica using a separate notebook command line. Do not edit and re-execute the same line. This way, your Mathematica notebook will keep a record of your data point entries. Save the notebook often.

Enter a data point into Mathematica using the *Mossbauer Helper* **`bkwd[ ]`** and **`fwd[ ]`** functions. The arguments to these functions are *counts*, *time*. Include a decimal point with the time. Executing the function will add the point to the data set and print a table of the points in the set. To suppress the table output follow the function with a semicolon '`;`'. Figure 14 shows some example entries.

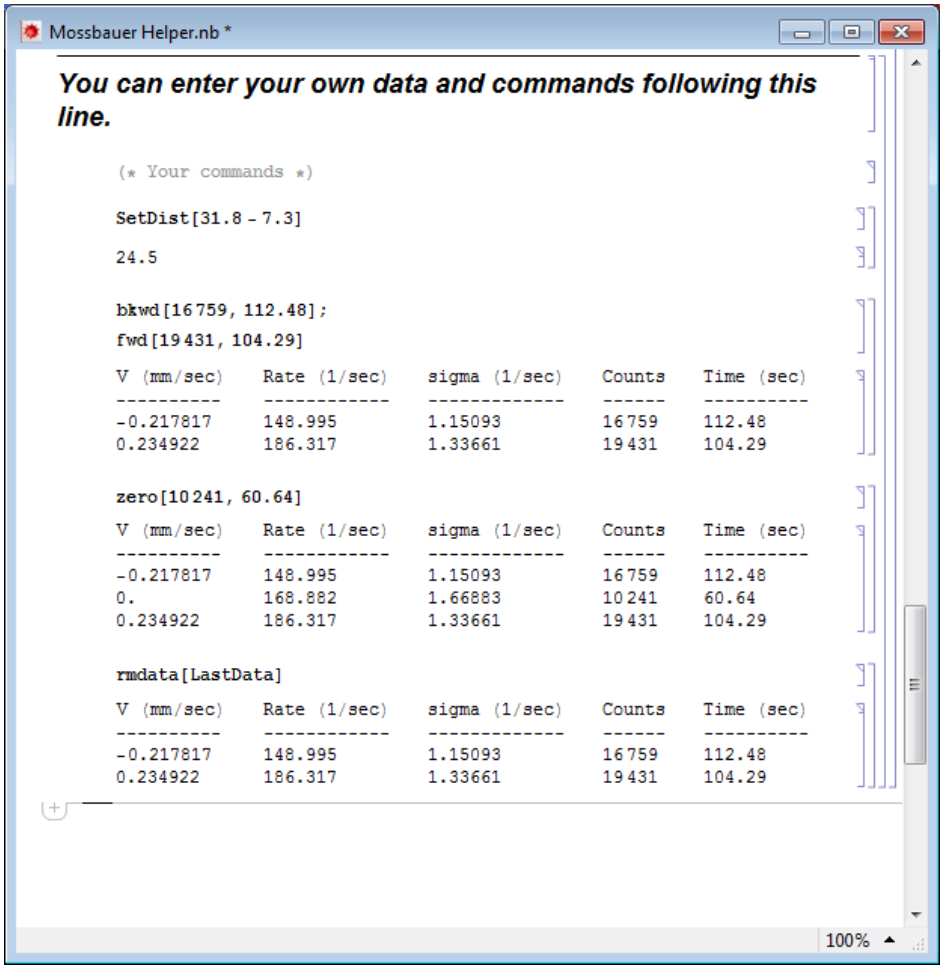


Figure 14: The first few data point entries showing how to use the *Mossbauer Helper* commands. The `SetDist[]` function should be the first one executed so that absorber velocities are properly calculated. `bkwd[`*counts*`,`*time*`]` and `fwd[`*counts*`,`*time*`]` will be the most often used functions. Use `zero[`*counts*`,`*time*`]` to enter a data point taken with the absorber motionless. Use `rmdata[]` to remove an incorrectly entered data point.

Taking a zero-speed point

The only absorber velocity which you can exactly repeat is with the absorber speed equal to 0 (motionless). Because zero absorber velocity is a repeatable data point, taking it every 20 minutes or so will let you check if the system count rate is unacceptably drifting. To take this point, do the following:

1. Set the motion select knob to **REST**.
2. Set the **AUTO**/**MNL** switch to **MNL** (manual), and the switch below it to **OFF**.
3. Reset both gate timers and both event counters.
4. Set the switch below the **AUTO**/**MNL** switch to **ON**. The timers and event counters should begin recording.
5. After a minute or so, return the switch below the **AUTO**/**MNL** switch to **OFF**. The timers and counters should stop.
6. Return the **AUTO**/**MNL** switch to **AUTO**.
7. Enter the data point into Mathematica using the *Mossbauer Helper* **`zero[ ]`** function.

Securing the apparatus

To properly secure the apparatus at the end of the experiment, do the following:

1. Remove the stainless steel absorber and the x-ray filter from the carriage and set them aside.
2. Turn off the power switch on the Mode Controller. Set the motion select knob to **REST**.
3. Set the temporary lead shield on the carriage between the source and the detector.
4. Remove the high voltage from the detector using the switch in the lower-left corner of the HV Power Supply. Leave the power supply power switch on.
5. Turn off the MCA using its **INTENSITY** knob.
6. Turn off the oscilloscope.

DATA ANALYSIS

Use *CurveFit* to fit the absorption line data to a Lorentzian+constant. Is this model for the line shape a good one? By what fraction is the count rate reduced at the center of the line? What is the relative isomeric shift of the line? Is the ^{57}Co more likely to be embedded in Rh or Pd?

Is the FWHM of the absorption line what you expect given the 98.1 ns half-life of the 14.4 keV state? Stainless steel is a mixture (alloy), not a compound. How might this affect the isomeric shifts of the individual ^{57}Fe nuclei and therefore the observed overall width of the absorption line? To fit the data to a Gaussian+constant rather than a Lorentzian, modify the data so that the y values transform to, say, $c - y$ for some choice of the constant c (the default *CurveFit* Gaussian fits will only fit peaks in the data, not absorption dips). Is the transformed data better fit by a Lorentzian or a Gaussian? The FWHM of a Gaussian is $2.36\,\sigma$. How does this compare to the Lorentzian fit's FWHM?

Poisson count statistics

The uncertainties in the count rates generated by the *Mossbauer Helper* functions are based on *Poisson count statistics*.[14] Assume that you look at a stream of rare, completely independent, random events. If the average event rate is r, then the expected number of events to arrive during a time period of duration t is $\mu = r\,t$, but the actual number of events to occur during any particular such time interval will vary randomly. The distribution of the numbers of events to occur is the *Poisson distribution*. Its standard deviation is equal to the square root of its mean, $\sigma = \sqrt{\mu}$. The *Mossbauer Helper* notebook makes an assumption often used when counting events: that $\sqrt{\mu}$ is reasonably well approximated by taking the square root of the actually observed count number. If the observed count number is n and the time interval is t, then the count rate and its uncertainty are estimated to be $(n \pm \sqrt{n})/t$.

Optional, for those more comfortable with Mathematica

The convolution of a Lorentzian and a Gaussian distribution is called a *Voigt distribution*.[15] The PDF of this distribution (as a function of x) may be defined in Mathematica using:

```
p[fwhm_, sig_, median_, x_]:= PDF[VoigtDistribution[fwhm/2,sig],x-median]
```

where **`fwhm`** is the Lorentzian's FWHM, **`sig`** is the Gaussian's standard deviation, **`median`** is the center of the distribution, and **`x`** is the location at which to evaluate the PDF, $p(x)$. This distribution may be a better model of the absorption line shape. See if you can use *CurveFit*'s *FitAnyFunction.nb* to fit your data using the Voigt distribution. What do the best-fit parameters tell you?

---

[14] After the French mathematician Siméon Denis Poisson (1781–1840).

[15] After the German physicist Woldemar Voigt (1850–1919).

PRELAB PROBLEMS

1. If radioactive ^{57}Co is embedded in a Rh matrix, then what would be the relative isomeric shift (in mm/sec) between it and the ^{57}Fe in a stainless steel absorber? Does this require a red-shift or a blue-shift of the 14.4 keV gamma ray photons from the ^{57}Co decay for absorption by the ^{57}Fe? What if ^{57}Co is in a Pd matrix?

2. The *half-life* of an excited state is the time it takes for its probability of remaining in that state to decrease by a factor of 2. Thus $\left|\langle\Psi_0|\Psi(t)\rangle\right|^2 \propto e^{-t/\tau} = 2^{-t/\tau_{1/2}}$, where τ is the state's *mean lifetime* and $\tau_{1/2}$ is its half-life. The 14.4 keV nuclear state of ^{57}Fe has a measured half-life of 98.1 ns. What is its mean lifetime τ? What is the expected FWHM (ΔE) of the state's Lorentzian energy distribution? What would be its equivalent Doppler velocity line width in mm/sec? When convolved with an absorption Lorentzian with the same line width, what should be the observed FWHM line width (in mm/sec) for the Mössbauer absorption line?

3. Derive the recoil energy equation (6), repeated below, relating the required incoming photon energy k to an absorber's nominal excited state energy E_a above its rest energy M:

$$k = E_a\left(1+\frac{E_a}{2M}\right)$$

4. Estimate the order of magnitude of the harmonic oscillator potential well energy $\hbar\omega_0$ (in eV) of an iron atom bound to its neighboring atoms in a crystal. Here's the logic: the angular frequency of the vibration mode of the harmonic oscillator is $\omega_0 = \sqrt{k_s/M}$, where k_s is the "spring constant" of the oscillator and M is the iron atom's mass. We know the atom's rest energy, so we need an estimate of k_s. The atom is bound to a few neighbors with chemical bonds of about an eV each, and pulling a pair of atoms apart by an angstrom or so will break the bond. The potential energy of the "spring" when stretched by this distance may then be roughly identified with the bond strength energy: $nE_{\text{bond}} \sim \frac{1}{2} k_s d^2$, where n is the number of bonds to the iron atom and d is the separation required to break a bond. Show that $k_s \sim 10\ \text{eV Å}^{-2}$. With the rest energy of the ^{57}Fe atom equal to 54 GeV and $\hbar c \approx 2000\ \text{eV Å}$, show that $\hbar\omega_0 \sim 10^{-2}$ eV.

5. *The Pound-Rebka experiment*: Einstein's general theory predicts that light will have its frequency shifted as it passes through a gravitational field. In an area with gravitational acceleration g (as near the Earth's surface), light will experience a gravitational red shift of

$$\frac{\Delta\omega}{\omega} = \frac{\Delta E}{E} = \frac{-gz}{c^2}$$

where z is the distance the light climbs against the gravitational acceleration g (it will be blue-shifted when falling in the direction of g). As mentioned earlier, Robert Pound and Glen

Rebka used Mössbauer emission and absorption of iron 14.4 keV photons to attempt to detect this shift. The source and absorber were separated by a vertical distance of 21 meters, and the absorption line's position was carefully measured with the absorber both above and below the emitter. The difference between the two absorption line positions was then identified with Einstein's predicted energy shift. Calculate the predicted energy shift of the 14.4 keV photons ($\Delta z = 2 \times 21$ meters) and compare it to the convolved Lorentzian absorption line FWHM you calculated in problem (2). Do you think that Pound and Rebka's result was a remarkable achievement given this comparison?