PHYSICS 359E: EXPERIMENT 2.2

THE MOSSBAUER EFFECT: RESONANT ABSORPTION OF (-RAYS

INTRODUCTION:

In classical physics resonant phenomena are expected whenever a system can undergo free oscillations. These occur with some characteristic frequency which is called the *natural* frequency. One example is the simple pendulum of length *l*, which has a natural frequency $\nu_0 = (1/2\pi)(g/l)^{1/2}$. Another is an electrical circuit consisting of an inductance L in series with a capacitor C which has a natural frequency $\nu_0 = 1/(2\pi(LC)^{1/2})$.

If a lightly damped resonant system is subjected to an external sinusoidal force at frequency ν, then the system will undergo forced oscillations with frequency ν and an amplitude proportional to

$$[(\nu-\nu_0)^2 + (\Delta\nu/2)^2]^{-1}$$

where $\Delta\nu$ is called the Full Width Half Maximum (FWHM) of the resonance, because the amplitude decreases by 1/2 when $\nu - \nu_0 = \pm\Delta\nu/2$. The FWHM is determined by the dissipation in the system: note that if $\Delta\nu$ were zero, the amplitude would be infinite when $\nu=\nu_0$. Physically, ν_0 represents the range of frequencies over which the resonant response of the system is close to its maximum. You should verify that the forced mechanical oscillator with small damping has exactly the response function given above with $\Delta\nu$ " the linear damping constant.

The relative width $\Delta\nu/\nu_0$ or its inverse, called the Q of the system, is used as a measure of the sharpness of the resonance. For simple mechanical systems, $\Delta\nu/\nu_0$ may have a value in the range 10^{-2} - 10^{-3}. For electrical circuits relative widths as small as 10^{-8} are possible, such as in a crystal-controlled oscillator.

In atomic systems, the resonant frequencies are associated with the energy differences between the allowed energy levels of the systems. For levels of energy E_i and E_j, the frequency ν_{ij} is given by the well-known Bohr relation

$$\nu_{ij} = * E_i - E_j * / h.$$

When an atomic system is subjected to an external force, the response is measured by the probability that the system will undergo a transition between allowed energy levels. Here we are interested in the special case of an atomic nucleus in its lowest (ground) energy level E_1 irradiated by photons of energy E. If the nucleus has an excited state of energy E_2, the probability that the nucleus will absorb a photon and be excited to the level of energy E_2 is proportional to

$$[(E - E_0)^2 + (' / 2)^2]^{-1}$$

where E_0 / E_2 - E_1. The FWHM of the resonance ' is related to the mean lifetime of the upper state by the uncertainty principle

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$$\Gamma \Delta t = \hbar / 2$$

Note again that ' is related to dissipation in the system: in this case the dissipative process is spontaneous emission which is responsible for the radiative decay of level E_2.

In atomic and nuclear systems '/E_0 may be very small. Usually it is less than 10^{-8} and may be as small as 10^{-15}. Thus the energy of an incident photon must be very close to the excitation energy E_0 if resonant absorption is to occur. At first sight, it might seem that photons emitted by the decay of nuclei originally in the excited state E_2 could undergo resonant absorption by other nuclei. In this radiative decay however, the nucleus must undergo a recoil in order to conserve linear momentum. Thus instead of emitting a photon of frequency $\nu = E_0/h$, the photon will have a frequency ν', which may be calculated as follows:

To conserve linear momentum the nucleus of mass M must recoil with a velocity V given by

$$h \nu' / c = M V.$$

Conservation of energy requires that

$$h \nu' + 1/2 \, MV^2 = E_0.$$

From these two expressions, we get

$$h\nu' [1 + (h\nu' / 2Mc^2)] = E_0 = h\nu.$$

If we assume that $h\nu'/2Mc^2 \ll 1$, then ν' is only slightly different from ν and we can set $h\nu'/2Mc^2$. $E_0/2Mc^2$. Thus the fractional change in frequency can be written as

$$(\nu - \nu')/\nu \ . \ E_0/2Mc^2.$$

In the particular case of interest to us, the first excited state of the nucleus ^{57}Fe (see FIG.1) has an energy E_2 = 14.41 keV (with E_1 / 0 keV). The lifetime of this level is known to be 9.78×10^{-8}s, so that ' = 1.1×10^{-27} J = 6.7×10^{-9} eV. Thus '/E_0 = 4.7×10^{-13}. On the other hand, the mass of ^{57}Fe is 56.935 amu (atomic mass units) or 5.34×10^{10} eV so that $E_0/2Mc^2$. 1.35×10^{-7}. It is clear that the energy shift arising from the nuclear recoil is very much greater than the natural width of the level. Thus the gamma rays emitted by a single nucleus will not undergo resonant absorption by other nuclei of the same type. By contrast, when an atom of comparable mass emits a visible photon (- 2 eV) from an excited level E_2 with a typical lifetime of 10^{-8} s, the recoil shift is much smaller than the natural width, so the photon can be absorbed by another similar atom in state E_1.

The above discussion assumes that the emitter and absorber are both at rest. In reality they are not, and their relative motion introduces a Doppler shift of the photon energy. For an ensemble of atoms, this shift appears as a Doppler broadening of the emission line, which usually dominates the natural

width. Even so, the recoil shift of the emitted (-ray or x-ray is usually greater than the Doppler broadening, making resonant absorption by a free nucleus impossible.

The Mössbauer effect refers to the emission of radiation from the nucleus of an atom bound in a crystal lattice. Under suitable conditions, the atom, with its nucleus, may remain fixed in the crystal lattice following emission of the photon. In this case, the recoil momentum is taken up by the whole crystal, rather than a single atom. The quantity M then refers to the mass of the crystal, and this will be greater than atomic masses by a factor on the order of Avogadro's number. If the resultant photon interacts with the nucleus of another atom similarly bound in a crystal lattice, the conditions for resonant absorption will be satisfied, and the probability is high that the photon will be absorbed. This resonant absorption as a result of recoiless emission and absorption of photons is known as the Mössbauer effect. See Ref.1, S2.4 for a discussion of how the probability for recoil-free emission or absorption depends on the Debye temperature of the crystal.

The Mössbauer effect is interesting in itself as an example of a resonant system. The importance of the effect in modern physics stems from the narrow width of the resonance. This means that the energy E_0 is very well defined, so that the effect can be used to study very small perturbations in E_0. One of the more spectacular uses of the Mössbauer effect has been to demonstrate the variation of photon energy in a gravitational field, in confirmation of a prediction of the theory of relativity.

The more usual cause of perturbations to E_0 is the presence of local fields within a crystal lattice, and the Mössbauer effect has been used extensively to study problems of crystal structure in solids. In this experiment, the Mössbauer spectra will show the effects of

a) isomeric shifts (Ref.1,S.4-4)
b) Zeeman splitting due to internal magnetic fields
c) interaction of a nuclear electric quadrupole moment with the gradient of the local electric field.

See the list of references for some guidance in researching these topics.

APPARATUS:

To demonstrate the existence of resonant absorption, it is necessary to be able to vary the photon energy by an amount comparable with the width of the resonance. This can be done through the Doppler effect. If a source of radiation of frequency ν_0 is moving with velocity v with respect to some observer, then the frequency measured by the observer is given by

$$\nu = \nu_0 [1 + (v/c)\cos 2]$$

where 2 is the angle between the velocity v and the vector from the source to the observer. To obtain shifts comparable to the width of the Mössbauer resonance, we require that

$$v = c(\nu - \nu_0)/\nu_0 = c\,'/E_0 \;.\; 0.1 \text{ mm/sec.}$$

Thus to vary the frequency ν at a constant rate, we must provide a constant acceleration to the source. The radiation source is ^{57}Co in a copper foil, which gives rise to the ^{57}Fe (-rays of interest

(see the decay scheme of ^{57}Co in Fig.1). The source can be mounted on a variable speed mechanical drive, which provides a controllable variation of photon energy. The (-rays are incident on an absorber containing ^{57}Fe and the transmitted ('s are detected in a proportional counter.

To collect a Mössbauer spectrum of (count rate vs. velocity (i.e. (-ray energy), the computer operates as an MCA in MCS mode. The synchronization of MCS channel advance and velocity increment is accomplished as follows (see FIG.2). An external oscillator produces pulses with a period)t (the dwell time) which are fed to the computer to advance the channel in which detector pulses are stored. These same pulses are sent to counter in the transducer control box, which thus produces a single square wave cycle for 1024 pulses. This square wave is integrated to produce a triangular ramp which is fed to the transducer to produce the desired velocity change. Thus each pulse from the oscillator increments the velocity by a fixed amount and causes counts to be stored in a new channel for a time)t.

To calibrate the speed of the transducer as a function of channel number, a Michelson interferometer using a He-Ne laser as a light source is set up with its moving mirror mounted on the transducer (see FIG.3). Every time the mirror moves by one-half of a wavelength 8/2, the interferometer light detector records one sinusoidal "fringe", which is amplified and converted to a narrow pulse by a zero-crossing detector. These pulses are fed into the computer in place of the counts, and counted for one dwell time)t. Since the number Ni of such pulses accumulated in channel i in one transducer cycle is given by

$$Ni = 2v_i)t/8 \text{ (prove this!)},$$

the velocity v_i for each channel can be calculated. This technique is described in Ref.4.

To carry out measurements you should familiarize yourself with the operation of the mechanical drive unit, and with the proportional counter and its associated electronics. It is prudent to turn off the Mössbauer drive unit before stopping the oscillator to avoid driving the shaft suddenly to one end. The ^{57}Co source is relatively strong, and will be handled only by the instructor. Relevant information is provided in the Radiation Safety Handbook available in the laboratory.

PROCEDURE:

1. Observe the (spectrum of the source with the computer MCA in PHA mode. You will need to isolate the pulses arising from the 14.4 keV (-ray by setting upper and lower limits on a SCA. You will find instructions for this in the write-up for the NUCLEAR LIFETIMES experiment, under "Setting up the SCA's".

2. With the experiment configured as in FIG.2, observe the Mössbauer spectra of the several absorbers which are available:

a) Sodium ferrocyanide. The internal fields in this material are negligible, and its absorption spectrum consists of a single line of width -2'. See Ref.1, S.3-52 for a relevant discussion of experimental linewidths.

b) Stainless steel. Small fields in the crystal lead to a broadening of the line.

c) Sodium nitroprusside. This molecule has an unsymmetrical structure which results in an electric field gradient at the location of the nucleus. The excited state of ^{57}Fe possesses an electric quadrupole moment which interacts with the field gradient to give rise to a level splitting.

d) Iron. Strong local magnetic fields exist at the site of the nucleus, leading to a Zeeman splitting of levels, and a consequent splitting of the absorption line. Close inspection of the differences in the transition energies will show evidence for quadrupole splitting as well.

3. Calibrate the transducer velocity (and hence the (energy) as described above.

After you have found energies for the observed transitions, you should compare them with theoretical expressions that take into account Zeeman and electric quadrupole effects. A least-squares fit will yield values for the physical parameters such as the product of magnetic moment and internal magnetic field.

REFERENCES:

1. H. Frauenfelder, *The Mössbauer Effect* (W.A. Benjamin, New York, 1962). The first five chapters present an excellent discussion of the theory and experimental techniques. It also contains reprints of many important papers in the field, i.e. Mössbauer's original publication (p.121); nuclear Zeeman effect(p.225); electric quadrupole effects(p.229); gravitational red shift (p.240).

2. L. May, *An Introduction to Mössbauer Spectroscopy* (Plenum Press, 1971). Ch.1 gives a good elementary discussion of theory. Ch.2 discusses experimental techniques. The remainder is detailed discussions of a variety of applications.

3. C.P. Slichter, *Principles of Magnetic Resonance*, 3rd or earlier ed. Has theory of electric quadrupole splitting in the presence of large and small magnetic fields.

4. G.M. Bancroft, *Mössbauer Spectroscopy* (Wiley, 1973). A very good treatment of the application of the effect to chemical problems.

5. Melissinos, p.256-79.

6. The writeup for the experiment on the Zeeman Effect (and references therein) contains information on Zeeman splitting in atoms. Note that Zeeman splitting of nuclear energy levels is simpler than in atoms because only one magnetic moment is involved, as in the case of the "normal" Zeeman effect in atoms.

7. Elscint instruction manuals, available in the lab.

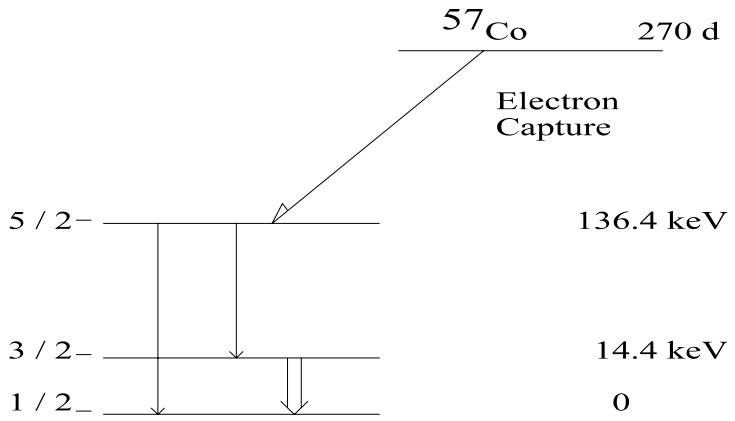

FIG.1. Decay scheme of %'Co

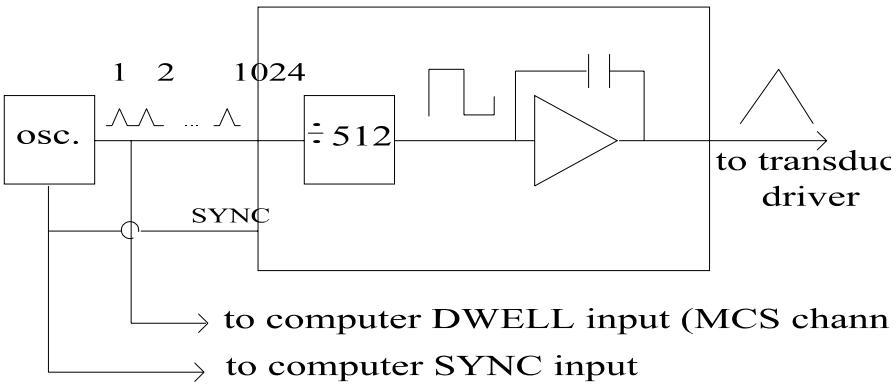

FIG.2. Ramp generator for transducer drive.

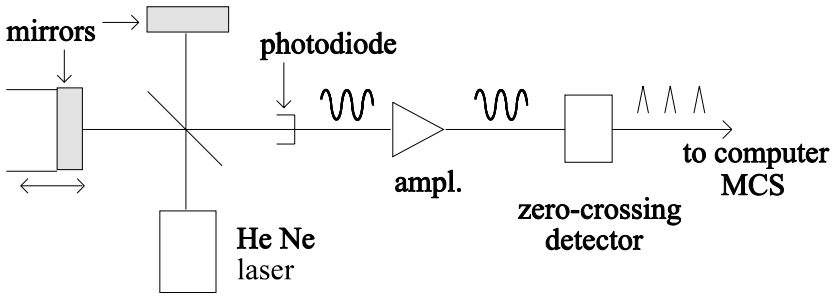

FIG.3. Velocity calibration with Michelson interferometer.