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Zutreffendes bitte ausfüllen

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1. Theory & Preparation

1.1 Resonant absorption & Mössbauer effect

The process of **resonant absorption** in nuclear physics describes the phenomenon of subsequent de- and excitation of two equal atoms to the same energy levels via one γ -quant. Consider for example the radioactive decay of ^{57}Co . Via electron capture it transforms into an excited state of ^{57}Fe , which in turn emits a 14.4 keV photon upon deexcitation.



In principle, due to the symmetry of the latter transition, one could use this emitted photon to raise another ^{57}Fe atom to the excited state. The photon is absorbed resonantly by the second atom during this process.

In reality, resonant absorption such as the Na-D-line only occurs under certain circumstances. Due to conservation laws the energy E_γ of the emitted photon does not exactly equal the transition energy E_0 , but is instead shifted downward by the nuclear recoil energy. A similar analysis finds that the energy for absorption of the same atom is shifted upwards.

$$\underbrace{E_\gamma = E_0 - \frac{p_\gamma^2}{2m}}_{\text{Emission}} \qquad \underbrace{E_\gamma = E_0 + \frac{p_\gamma^2}{2m}}_{\text{Absorption}} \qquad (1.1)$$

With the photon impulse p_γ and atom mass m . If the line width introduced by natural broadening or other effects does not exceed the energy gap, resonant absorption cannot occur (see Figure 1.1). It is also notable that the energy gap between emission and absorption spectrum can be increased by additional effects. This will be further discussed in chapter 2.

As it turns out, the above rules stating when resonant absorption can and cannot occur are not strictly true. Experiments in the 1960s conducted by R. Mössbauer ([Wer13]) showed that resonant absorption in a crystal lattice happens much more readily than one would expect based on the previous discussion. The difference is the tight binding of the atoms in the crystal lattice. Instead of the individual atom recoiling, different phonons can be created (or destroyed) by the emission and absorption. In a sense, the entire crystal absorbs the recoil energy, effectively substituting the atom mass m in equation Equation 1.1 by the mass M of the entire crystal. Because $m \ll M$, the energy gap between emission and absorption spectrum drastically decreases. This phenomenon of recoilless nuclear resonant absorption is named **Mössbauer effect**.

1.2 Mössbauer spectroscopy

To measure the the natural linewidth of ^{57}Fe with roughly 5 neV common measurement methods will fail. Even with high-resolution interferometers, some orders of magnitude

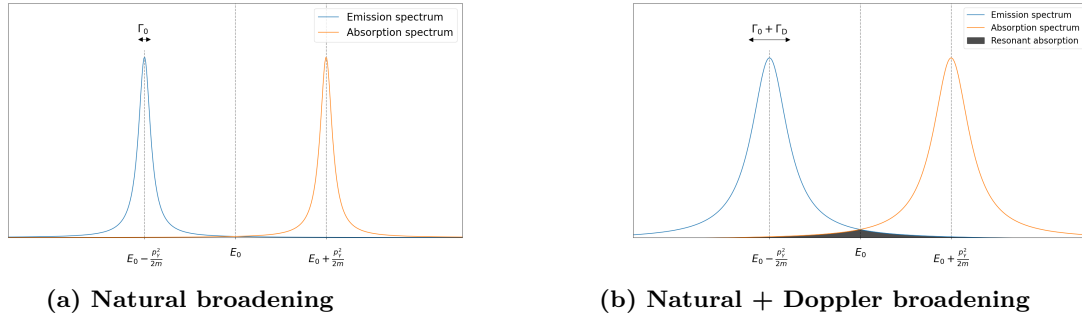


Figure 1.1: (a) The natural linewidth Γ_0 is not sufficient for a sizeable overlap of both spectra. Resonance absorption is not possible. (b) The line width of both spectra can be increased by other effects such as Doppler broadening. In such cases the spectra with linewidth $\Gamma_0 + \Gamma_D$ can overlap and resonant absorption is possible.

are still missing to resolve such lines by direct measurement of the spectrum. Therefore we take the Mössbauer effect into consideration. It is possible to measure the resonant absorption by the transition rate of the photons. The detector does not need to be energy sensitive, it is sufficient to detect the quanta in this case. The magnitude of the transition displays the overlap of the probabilities for emission and absorption (see Figure 1.1) of the natural lines. By varying the distance of the emission and absorption peaks we can make a statement about the line profile.

If the gamma radiation source is in motion a **Doppler shift** occurs and the photon energy changes. Therefore we can slightly modify the photon energy by changing the velocity of the source (see Equation 1.2).

$$\Delta E_\gamma = \pm \frac{v}{c} \cdot E \quad (1.2)$$

With the measured transmission spectrum as a function of the velocity of the source one can make qualitative as well as quantitative statements about the element. Furthermore it is possible to observe three types of nuclear interaction: isometric shift, quadrupole splitting and hyperfine magnetic splitting. The FWHM of the transmission curve corresponds to the double natural linewidth of the transition.

2. Experiment & Evaluation

2.1 Experimental setup

The experiment consists of several components. A schematic setup can be seen in Figure ???. The γ -source is moving at a velocity relative to the lab frame. The velocity can be adjusted via a digital function generator (DFG), Mössbauer velocity calibrator (MVC), Mössbauer velocity transducer (MVT), and Mössbauer driving unit (MDU). An absorber can be placed in the beam path of the high-energy photons. Based on their energy, the γ 's are either transmitted or absorbed by the target. The number of transmitted photons is counted by a 1024 channel multi-channel-scaler (MCS). The various other components of the setup (VV, etc.) are used mainly for data acquisition (DAQ) purposes, more detailed information on the individual building blocks can be taken from [Sch17].

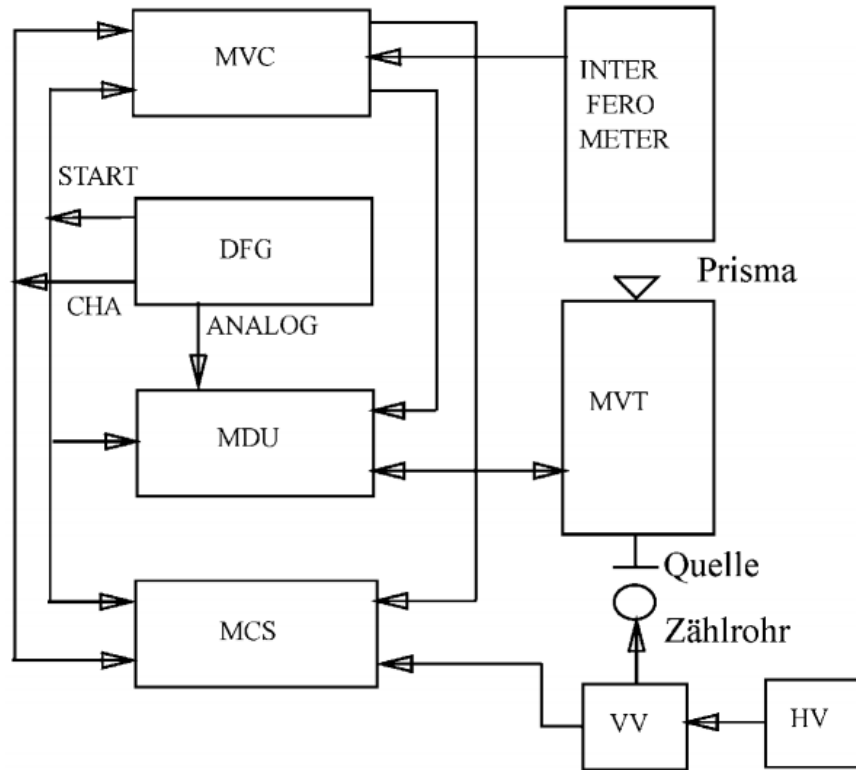


Figure 2.1: A schematic overview of the experiment components. The Mössbauer driving unit (MDU) moves the γ -source at a velocity relative to the multi-channel-scaler (MCS). The exact velocity can be controlled via a digital function generator (DFG), Mössbauer velocity transducer (MVT) and is monitored by an interferometer.

2.2 Measuring the 14.4 keV ^{57}Fe -line

As described in section 1.2, Mössbauer spectroscopy does not rely on an extremely precise measurement of the photon energy. Due to resonant absorption it is sufficient to merely count the number of photons transmitted through an absorber to gather information about its atomic energy levels and existing nuclear transitions. However, in order to allow for more detailed analysis in the following evaluation the relative difference of transition energies needs to be quantified. For this purpose as well as a proof of concept, the γ -spectrum of ^{57}Co is measured. Visible in the observed spectrum in Figure 2.2 is a characteristic Lorentzian peak that represents the 14.4 keV resonance of the excited $^{57}\text{Fe}^*$ state. The exact fitfunction as well as fit results are given in Equation 2.1. The propagated uncertainty in f is also given.

$$f(\mathcal{C}, A, \omega_0, \gamma) = \frac{A}{(\mathcal{C}^2 - \omega_0^2)^2 + \gamma^2 \omega_0^2} \quad (2.1)$$

$$A = (8.68 \pm 0.36) \times 10^{10} \quad (2.2)$$

$$\omega_0 = 67.84 \pm 0.08 \quad (2.3)$$

$$\gamma = 23.25 \pm 0.52 \quad (2.4)$$

$$\text{COV}(A, \omega_0, \gamma) = \begin{bmatrix} 1.28 \times 10^{19} & 1.00 \times 10^7 & 1.86 \times 10^9 \\ 1.00 \times 10^7 & 5.66 \times 10^{-3} & -6.41 \times 10^{-4} \\ 1.86 \times 10^9 & -6.41 \times 10^{-4} & 2.75 \times 10^{-1} \end{bmatrix}$$

$$\Delta f(\mathcal{C}) = \sqrt{(\nabla f(\mathcal{C}))^T \text{COV}(A, \omega_0, \gamma) \nabla f(\mathcal{C})}$$

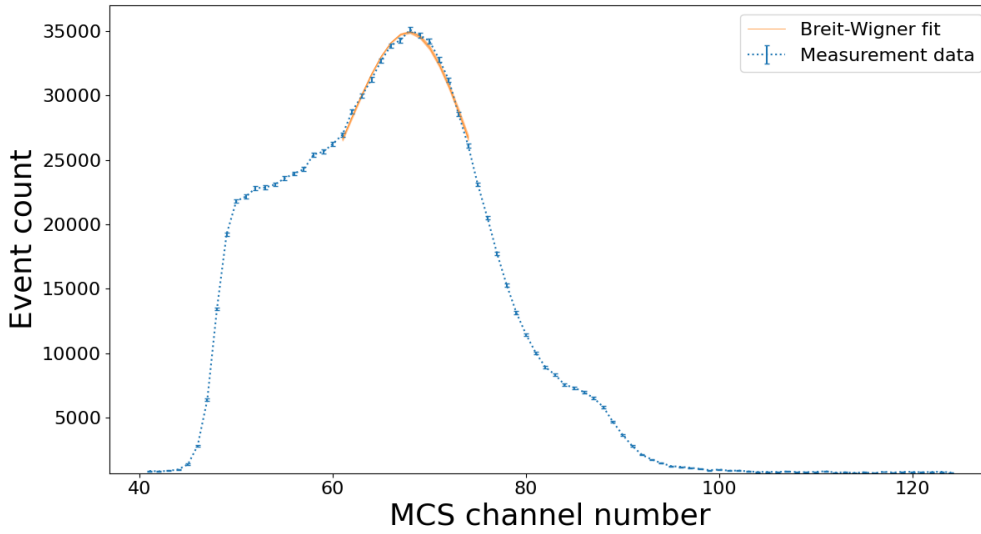


Figure 2.2: The error on the measured counts is assumed to be Poissonian. Because the amplitude A does not hold physical information other than the flux of the γ -source, uncertainties and correlations in A are neglected in the below error propagation.

2.3 Measuring the Mössbauer spectrum

In the following section the Mössbauer spectrum of various materials is measured. The process of how data is collected and analysed is presented using the example of an iron absorber. Other materials are analysed in a similar fashion.

2.3.1 Iron

The iron absorber is placed in the beam path. The transmitted photons are counted for roughly 20 min. In this measuring mode, the MCS channel number corresponds to a velocity v at which the γ -source is moving relative to the iron target. The 1024 channels available for readout are split into two 512-channel intervals (in the following addressed as Ch1 and Ch2) that differ in the acceleration of the source. It is assumed that the measurements of Ch1 and Ch2 respectively are uncorrelated. As lined out in the lab manual [Sch17] it is assumed that the maximum velocity $v_{\max} = \pm 10 \frac{\text{mm}}{\text{s}}$ corresponds to the edges of the channel intervals (i.e. Channel #0 and #1023 for positive velocity, channel #511, and #512 for negative velocity). Using this information, a relation between channel number \mathcal{C} and γ -source velocity can be constructed as follows:

$$v(\mathcal{C}) = 10 \frac{\text{mm}}{\text{s}} \cdot \left(\frac{\mathcal{C} - 256}{256} \right). \quad (2.5)$$

Because of this relative velocity the photons that are emitted at an energy of 14.4 keV are Doppler shifted to slightly lower/higher energies. If now a nuclear transition from state $|i\rangle$ to state $|f\rangle$ exists for an iron atom in the crystal lattice where $E_f - E_i = E'$, there is a nonzero probability that the atom absorbs the photon and transitions to the higher energy state. Consequently, a dip in the photon spectrum at that specific energy (and by extension a specific velocity) can be observed. The resonance around this energy E' can be modelled by a slightly modified Breit-Wigner shape presented in Equation 2.6.

$$N(v) = \Phi_0 - \frac{A}{(v - v_0)^2 + (\frac{1}{2}\Gamma)^2}, \quad (2.6)$$

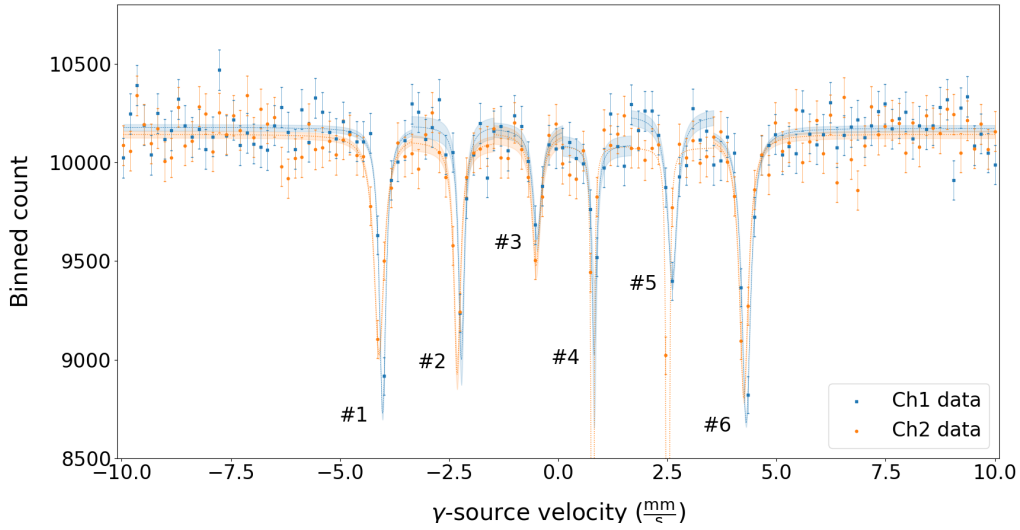


Figure 2.3: Mössbauer spectrum of iron

Six absorption peaks can be seen in the Mössbauer spectrum of iron. They are labelled #1 to #6. The two measurement channels are colour-coded respectively.

where Φ_0 is the integrated γ -flux (i.e. number of photons with energy $E \approx 14.4 \text{ keV}$). The normalisation factor A , v_0 the velocity of the γ -source at which the photons are shifted by the transition energy E' and Γ the full-width-at-half-maximum (FWHM) value of the absorption peak. Technically, using this parametrisation of the Breit-Wigner curve, Γ should also appear in the numerator. To ensure a more stable fit result, it has however been absorbed in A .

Fitting Equation 2.6 to measurement data, six absorption peaks can be identified (see Figure ??) depending on the velocity of the γ -source. The individual parameters optimised to model the observed spectrum are listed in Table 2.1. It is important to note that absorption peak #4 and #5 are not properly fitted for data from Channel 2. The information gathered from these peaks will be ignored in the following analysis. For every other absorption peak the corresponding fit parameters P will be combined to a mean value \bar{P} .

$$\bar{P} = P_{\text{Ch1}} + P_{\text{Ch2}}$$

$$\Delta \bar{P} = \sqrt{\Delta P_{\text{Ch1}}^2 + \Delta P_{\text{Ch2}}^2}$$

Due to the use of different emitter and absorber materials, a **isomer shift** occurs. This is caused by the different charge radii and electron distribution of the states involved in the transition. In the Mössbauer spectrum this results in a fixed shift of all absorption peaks around the center ($v=0$), which overlaps with other phenomena. The isomer shift can be calculated by determining the center of gravity of pairwise connected lines.

At the ^{57}Co -Rh source with natural iron as absorber an isomer shift of

$$\Delta E = (1 \pm 2) \cdot 10^{-9} \text{ eV} \quad (2.7)$$

occurs.

Table 2.1: Mössbauer spectrum fit parameters for iron

Peak #	Φ_0	A	v_0	Γ	Channel
#1	10177 ± 16	12 ± 3.9	-4.02 ± 0.02	0.184 ± 0.043	Ch1
	10142 ± 17	21 ± 6.0	-4.11 ± 0.02	0.277 ± 0.049	Ch2
#2	10187 ± 53	8 ± 6.2	-2.22 ± 0.03	0.161 ± 0.104	Ch1
	10108 ± 38	10 ± 5.6	-2.32 ± 0.01	0.184 ± 0.080	Ch2
#3	10173 ± 37	8 ± 5.2	-0.49 ± 0.02	0.234 ± 0.099	Ch1
	10151 ± 47	9 ± 5.6	-0.51 ± 0.03	0.235 ± 0.076	Ch2
#4	10092 ± 50	4 ± 6.0	0.82 ± 0.02	0.130 ± 0.193	Ch1
	10082 ± 65	2 ± 6.1	0.79 ± 0.05	$0.000 \pm \infty$	Ch2
#5	10243 ± 38	15 ± 5.4	2.61 ± 0.02	0.266 ± 0.048	Ch1
	10075 ± 24	3 ± 2.4	2.51 ± 0.02	$0.000 \pm \infty$	Ch2
#6	10173 ± 17	23 ± 5.1	4.30 ± 0.01	0.246 ± 0.036	Ch1
	10142 ± 19	21 ± 6.9	4.25 ± 0.01	0.250 ± 0.058	Ch2

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