

PALACKÝ UNIVERSITY OLMOUC
FACULTY OF SCIENCE
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MASTER THESIS

Development of a spectrometric chain for
the detection of low-energy gamma
radiation using semiconductors.



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DECLARATION

I hereby declare that I elaborated this bachelor thesis independently under the supervision of Mgr. Aleš Stejskal Ph.D., using only information sources referred in the Literature chapter.

In Olomouc December 26, 2023

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Introduction

Chapter 1

Gamma rays properties and matter interaction

1.1 Gamma emission

In opposite to alpha and beta particles, the gamma rays are without charge and their rest mass is zero, and thus they are considered to be high-energetic photons, but the mayor difference to the RTG photons they originate only from atomic nucleus upon its deexcitation from higher energy level to lower. The energy levels of nucleus are similar to the levels in electron shell - discrete, characteristic for every isotope and can be described by quantum numbers. The gamma emission is usually follows alpha or beta decays. The gamma photon is emitted, because the new nucleus is created in an excited state.

1.2 Passage of radiation and particles through matter

Interaction of a particle (radiation) with another particle (atom, nuclei, free electron) or with continuous matter can result into many types of reactions and effects - scattering of a particle from incident direction, creation of new particles and nuclei, annihilation of incident particle etc. It mainly depends on particle's energy, electric charge, spin and mass, but also on the properties of target particle or matter. The physical quantity describing the probability of a specific interaction of particle with single point target is known as the cross section. Normalized to the unit solid angle - differential cross section:

$$\frac{d\sigma}{d\Omega} = \frac{1}{F} \frac{dN_s}{d\Omega} \quad (1.1)$$

Where F is a particle flux, Ω is a solid angle and N_s is the average number of scattered particles per unit time. And the total cross section is given by integration:

$$\sigma = \int \frac{d\sigma}{d\Omega} d\Omega \quad (1.2)$$

However, to characterize the interaction probability of particle with continuous matter, which contains many interaction centres (defined by their density), other assumptions have to be made. The average number of scattered particles is given by:

$$N(\Omega) = FAN\delta x \frac{d\sigma}{d\Omega} \quad (1.3)$$

and integrated:

$$N_{tot} = FAN\sigma\delta x \quad (1.4)$$

The A is a total area perpendicular to the flux, δx is the material thickness and N is the density of interaction centres.

Heavy charged particles (such as alpha particles, protons, muons, pions) lose their energy mainly due to the atomic electrons collisions. Due to their mass which is much higher than the mass of electrons ($M \gg m_e$) they collide with, the direction of their path is left unchanged. The loses of energy per unit path is defined as stopping power $\frac{dE}{dx}$. The stopping power for the heavy charged particles is given by Bethe-Bloch formula which relates stopping power and particle's energy. However the Bethe-Bloch formula doesn't apply on low energies (Lindhard-Sharf nuclear loses) and on higher energies (bremsstrahlung radiation). The change of their path direction is possible by the second process with lesser probability - by the elastic scattering from nuclei.

Electrons and positrons have much smaller mass than the heavy charged particles, and thus the direction of their path is changed due to the movement in an electric field of nucleus. The bremsstrahlung radiation loses are mayor yet at low energies. However, the energy lost due to the collisions also comes to play - it is guided by special Bethe-Bloch formula, which takes the path direction change into account.

Other interaction effects are also possible (Cherenkov radiation emission, nuclear reactions), but they are rare or does not affect the particle's energy as those previously mentioned.

The interaction of neutrons is totally different due to the lack of charge.

The interaction effects for gamma rays are described more detail in the following chapter.

1.2.1 Gamma matter interaction

Due to the fact, that the gamma rays are photons, the gradual losses of kinetic energy inside materials (characteristic for the charged particles) are not observed. Instead the main observed effect is the attenuation of intensity of photon flux with the increasing thickness of the absorber material.

Three mayor interaction effects of gamma photons and matter are Photoelectric effect, Compton scattering and Pair production. The cross section of these effect vary with gamma photon energy, with material and its density - high dependence on atomic number Z . There are also possible nuclear reaction such as the Mössbauer effect, but their observation requires very special conditions to be met.

The attenuation of of photon flux has a form of exponential decay:

$$I = I_0 \exp(-\mu x) \quad (1.5)$$

where I_0 and I are the intensities and the parameter μ is the total absorption coefficient, which describes the probability of interaction per unit length and is bounded with total cross section of single atom σ (combination of cross section of three main effects) by relation:

$$\mu = N\sigma = \sigma(N_a\rho/A) \quad (1.6)$$

where N - density of atoms, N_a - Avogadro number, ρ - material density and A - molecular weight.

The dependence of total cross section combined of the three main effect for lead on photon energy can be seen on fig. 1.1.

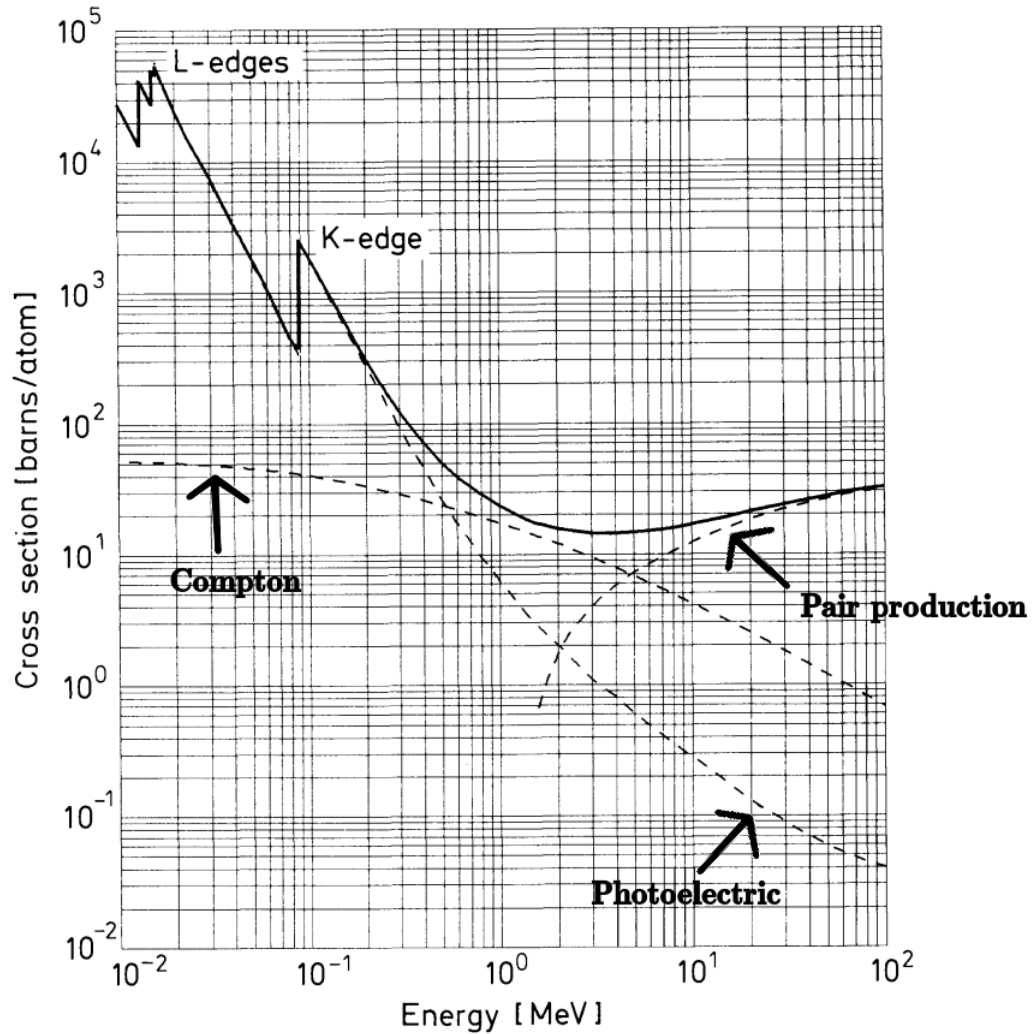


Figure 1.1: Total absorption cross section for high-energy photons. Taken and modified from [Leo1987-wy].

Photoelectric effect

The photon is absorbed by electron from atomic shell. The electron is ejected and acquires the kinetic energy given by well-known equation:

$$E = hf - E_b, \quad (1.7)$$

where f is the photon frequency and E_b is the binding energy.

The Photoelectric effect can be observed only on electron bounded in atomic shell, due to the fact, that the nuclei can absorb the photon's momentum. The cross section usually falls with energy and can have characteristic peaks from K,L,M transitions.

The photoelectric effect plays the key role when it comes to the detection of gamma photons.

Compton scattering

Compton scattering is an effect, which is mainly observed on free electrons. It needs to be said, that the electrons in material are usually bounded, however, if the photon energy is much higher than the binding energy

This effect causes, that the photon loses only a part of its energy and changes its direction. The energy is transferred to an electron accordingly to the laws of energy and momentum conservation.

Pair production

At much higher energies, the photon can be converted into electron-positron pair. This effect happens near the nucleus, which absorbs a part of the original photon momentum.

The pair production along with electron bremsstrahlung radiation are key effects for electron-photon shower development. If the created electron/positron has sufficient energy, it emits bremsstrahlung photons. These bremsstrahlung photons can again interact via the pair production, and thus creating new electrons again. This process of shower development continues until the energy of electron/positron pairs drops under the energy, which is required to produce bremsstrahlung radiation.

For low-energy gammas the cross-section is so small, which means that for the purposes of this thesis, this interaction effect could be neglected.

1.2.2 Characteristic energy spectra

Every gamma source emits its characteristic photons with certain probabilities. However, before these photons are captured by detector, many effects can occur in environment or inside the detector - photons can interact various materials without detection purpose such as shielding, spectrum can be altered by noise events in detector etc.

Chapter 2

Gamma rays detection

2.1 Properties and parameters of detectors

2.1.1 Gaseous detectors

2.1.2 Scintillation Detectors

2.1.3 Detectors based on semiconductors

Chapter 3

Semiconductor detectors with p-n junction

The semiconductor structures has unique properties, which make them usable not only for a particle/photon detection. There are many types of semiconductor detectors - conventional detectors of light intensity with spectral range from infrared to UV - photoresistors and photodiodes, low intensities and single photons detectors - avalanche photodiodes (APDs), for imaging - CCD and CMOS cameras, x-ray, gamma and other particles rates and energy detectors - special photodiodes, for particle position and energy measurements - matrix drift and strip detectors. The essential role for gamma spectroscopy have the p-n junction detectors - detection diodes capable of capturing gamma photons with sufficient energy resolution and efficiency.

To detect Mössbauer 14.4 keV gammas our primary choice are the Si PIN diodes for direct detection due to the guaranteed high detection efficiency under 25 keV.

3.1 The crystal structure of semiconductor

Since we are not dealing with single atoms characterized by the well-known discrete energy levels, but with solid crystals, there is another model describing the energy levels - the band structure. The energy levels are very close each other that they nearly form a continuum. This different behaviour is a result of a periodical potential inside the crystal lattice. However, the electron energy E_n is not the only quantum number describing the behaviour of electrons inside the periodical potential. By solving Schrödinger equation with the help of so-called Bloch's theorem, we come to a conclusion that the second important quantum number is the wave vector \mathbf{k} . These two quantum numbers are bounded together by dispersion relations $E = E(\mathbf{k})$, which is characteristic for every crystal and may play role when it comes to transitions of electrons between bands.

3.1.1 Band structure

The basic difference between conductors, insulators and semiconductors is the valence and conduction band structure. In case of conductors, the valence band overlaps the conduction band. The electrons in conduction band can move nearly freely throughout the crystal. For insulators, there is no overlap, and the energy gap E_g between the two bands is so high, that it makes any transitions of electrons nearly impossible. However in case of semiconductors, the E_g is small enough to allow the electron jump into the

conduction band and leaving the hole in valence band after receiving a sufficient amount of energy - in form of thermal energy $E_T = kT$, energy in static electric field $E_s = -e\phi$ or as a photon with energy quantum $E_{\text{photon}} = \hbar\omega$. The last one is the most important, because it allows us to use the semiconductors as detectors. Both the electron and hole participates on conduction, but they have to be treated as quasiparticles with their specific masses and mobilities. At $T = 0$ K, the electrons occupy the lowest possible states and the highest occupied state is defined by energy of Fermi-level E_F

3.1.2 Crystal lattice

The semiconductor materials have usually form of a crystal of diamond structure (two FCC lattices bounded together.) with the lattice atoms bounded by a covalent bond. However in real crystals there are also defects and impurities which may alter the functionality. The impurities can be added on purpose to enhance the properties we seek in procedure called doping.

3.1.3 Direct and indirect semiconductors

The shape of dispersion relations between energy and $E(\mathbf{k})$ divides the semiconductors into two categories - direct and indirect semiconductors. For direct semiconductors the minimum in conduction band and maximum in valence band have the same \mathbf{k} , so the transition can occur after the E_g is supplied. In case of indirect the minimum and maximum have different \mathbf{k} . To make transition, additional momentum has to be server (for example by phonons) or the supplied energy must be much larger to allow transition to the higher state. The Si crystal is an indirect semiconductor.

3.2 P-N junction

The intrinsic semiconductors have only a little field of applications.

The

- it could be modelled as a simple plate capacitor. The capacitance of p-n junction plays a significant role - it alters the dynamical parameters of the detection circuits - it can alter the rising edges of pulses, alter the entire frequency spectra, increase/decrease the SNR.

3.3 Fabrication of semiconductors

3.3.1 Materials

The suitable materials for construction of ionizing radiation detectors are Ge, Si, GaAs and CdTe. The CdTe is used in wide-spectre X-Ray and Gamma Ray Detector.

3.4 Detection mechanism

If we want to detect a particle or photon, it has to interact with the detector material - in semiconductor the creation of electron-hole pairs is observed. This could be achieved by interaction mechanisms described in previous chapters, which differ for every type of particle and energy.

The main of purpose of semiconductor as a detector, is to perform a linear conversion of the particle/photon energy into the free charge carriers - electrons in conduction band and holes in valence band. The average energy needed to create a single electron-hole pair is usually a fix constant - in Si it is $\epsilon = 3.6$ eV [Lutz²⁰⁰⁷]. Since the Si is an indirect semiconductor, this value is not equal to gap energy, which is lower ($E_g = 1.12$ eV). In case of low-energy gamma the two main mechanisms which take place in producing the charge carriers are photoeffect and compton scattering.

The gamma photon striking the detector firstly interacts with a single electron. If the photoeffect takes its place, the primary electron with energy much higher than the thermal excitation will ionize the electrons from all over the valence band pushing them into higher states from conduction band. These Electrons in conduction band deexcite themselves into lower states of conduction band and the holes redistribute themselves to the upper states in valence band. This redistribution process releases energy again which leads to the cascade of further excitations and interactions, which produce many electron-hole pairs - the charge cloud. Number of generated electron-hole pairs n is simply given by relation:

$$n = \frac{E_{\text{gamma}}}{\epsilon}. \quad (3.1)$$

In case of compton, the scattered photon may escape the detector without any other interaction taking the rest of its energy out of the detector. This results into energy distortion. The compton without any other interaction (compton again or photoeffect) is typical for thin detectors since there is insufficient thickness to stop the scattered photon again. In detected gamma energy spectrum this leads into compton continuum with the compton edge, which devaluates the spectra.

An ideal very thick detector could solve this problem, because every possible interaction and all the following sub-interactions will happen inside the detector and will result into electron-hole generation. The detector of this kind would have only the full energy peaks without any other unwanted counts. However, construction of such detector is impossible due to the many technical and manufacturing problems arising with increasing dimensions.

In case of p-n junction the created charge carriers in depleted layer are pushed towards electrodes by internal electric field. The electronics accumulates the charge and converts it into the voltage pulse signal, which is then analysed to extract the energy information.

3.5 Main noise sources and resolution limitations

The detection efficiency and the energy resolution are strongly dependent on noise which alters the signal. While some negative effects are caused by outer conditions such as temperature or by noise in electronics, some originate strictly from semiconductor material properties and only chance to minimize them is during the crystal's preparation.

3.5.1 Fano noise

The physical effect inside the semiconductor itself which limits the measured energy resolution is the fact that not all of the particle's energy is transformed into charge car-

riers. Some fraction of energy can be consumed to induce lattice vibrations (phonons). Statistically the relative resolution can be described by following equation:

$$\frac{\Delta E}{E} = 2.35 \sqrt{\frac{F\epsilon}{E}}, \quad (3.2)$$

where E is incoming particle's energy, ϵ is the average energy needed for the single pair creation and F is a Fano Factor. Fano Factor is a special statistical constant which describes the dispersion in counting process. For Poisson distribution the F is 1. If we consider the case of 14.4 keV gamma inside Si ($\epsilon = 3.6$ eV, $F = 0.12$), the $\frac{\Delta E}{E}$ is equal to 1.3 % and ΔE is 185.4 eV.

This resolution is sometimes called intrinsic, because the real resolution is much worse. This model does not consider other sources of noise coming from the amplification in electronics, dark currents e.g.

3.5.2 Thermal noise

It was mentioned that the electrons can be also excited by thermal fluctuations. However, since the many of detector's materials are indirect semiconductors, the thermal excitations are carried through the intermediate states inside the band gap. These states have the origin in imperfections and impurities inside the crystal lattice. The electrons and holes generated this way can join the charge cloud generated by detected particle and decrease the SNR. This phenomenon is usually called dark current.

The thermal fluctuations are much more significant for materials which require less energy to excite the electrons to the conduction band such as Ge. These detectors always have to be operated at low temperatures. Even the room temperature can lead up to the breakdown and destruction of a detector. For Si, the valence and conduction band are more distant. so the thermal fluctuations are not so critical.

3.5.3 Recombination and trapping

The effect which goes against the successful collection of charge carriers and thus affects the detection efficiency and the energy resolution is recombination. In pure crystals, the electron can recombine with hole only when they have the same \mathbf{k} . However, inside real crystals there exists impurities, which allows carriers to recombine - recombination centres. The charge collection must take much less time than it takes the carriers to recombine.

Another effect caused by the very same impurities is trapping. The trapping centres capture electron or hole and release it after certain time. This effect alter the signal pulses and can decrease maximum possible counting rate.

Energy levels near the centre of band gap are primary responsible for recombination and trapping, which means that the impurities causing this are not the impurities which were added in doping, because these have their additional energy levels near the bands.

3.6 Construction scheme of Instruments for gamma detection based on semiconductors

3.6.1 XR-100CdTe X-Ray and Gamma Ray Detector

3.6.2 MIMOS 2

3.7 Detector structure

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Its obvious that for particles' interactions inside the detector the most crucial layers' dimensions are those in cross section normal to the incident radiation. The dead layer (usually the p) has to be thin as possible to reduce probability that the particle interacts in place where the generated charge cloud would nearly immediately recombine. The active layer intrinsic

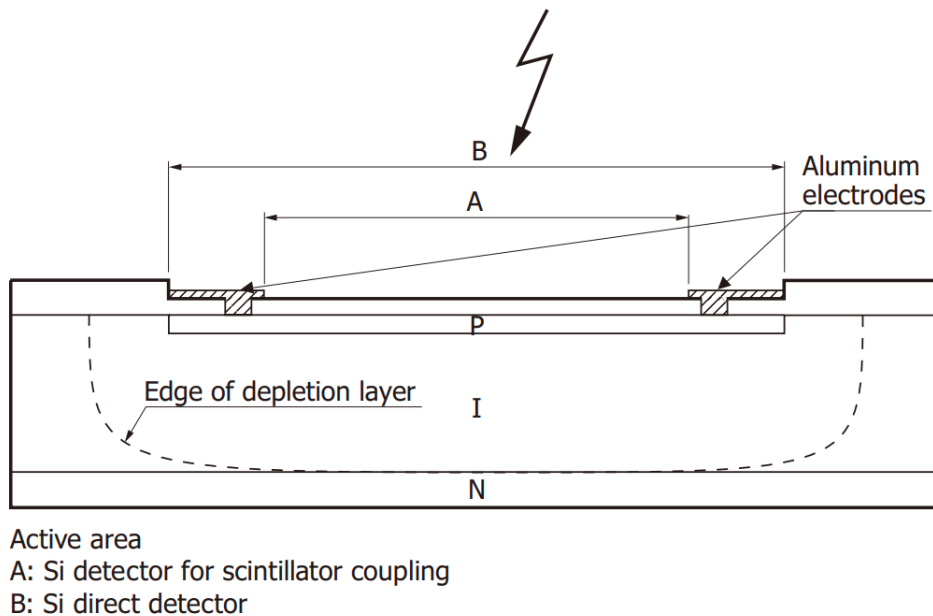


Figure 3.1: Cross section of Si detector for scintillator coupling and Si direct detector [SiPINdirect].

3.8 Available detection diodes

For testing we have chosen one professional Hamamatsu s14605 PIN diode and two commercial PIN diodes: OPF420 - originally designed to be used in optical fibres, BPW34 - visible and near infrared radiation detector.

3.9 Hamamatsu s14605

3.10 OPF420

3.11 BPW34

Chapter 4

Electronics for signal readout and analysis

4.1 Diode detector connection and support electronics

4.1.1 High voltage source

4.1.2 Shielding and grounding

4.1.3 Cooling

To reduce thermal noise and achieve better SNR it is necessary to cool the detector.

4.2 Spectrometric chain

4.2.1 Pre-amplification

4.2.2 Amplification and shaping

4.2.3 Pulse-height measurement and spectra accumulation

Chapter 5

Mössbauer effect and spectroscopy

Mössbauer effect is a physical effect, which can under certain circumstances occur on atomic nuclei. It consists of recoilless resonance emission/absorption of gamma photons by nuclei of source/absorber with discrete nuclear energy levels. This effect has a wide field of application - mainly the Mössbauer spectroscopy, which is a nuclear experimental technique and a special type of gamma spectroscopy, which uses the appropriate nuclei in studied sample as sonds of local electric and magnetic fields.

This technique is capable of providing many unique information in material research, geology, chemistry and biology - study of phase and chemical composition of solid materials such as steel, study of local magnetic fields and spin states, in-situ measurements of phase transitions. The main disadvantage of this technique is the fact that there is not an appropriate radiation source for many isotopes. The mayor significance has iron and its isotope ^{57}Fe with possible radiation source ^{57}Co (which decays into an excited state of ^{57}Fe), which allows Mössbauer spectroscopy to be employed on many fields, including the steel industry.

5.1 Physical concept

5.1.1 Resonance emission and absorption

In previous chapters was mentioned, that the atomic nuclei are quantum systems with discrete energy levels (analogous to the energy levels of electron shell), thus upon deexcitation or excitation they emit/absorb gamma photon with energy E_0 equal to the difference between the levels. For the free, stationary nucleus, the emitted/absorbed energy spectra follow the shape of Lorentzian curve.

However, this energy may be altered - due to the momentum conservation law, some part of the gamma photon energy is transferred to the kinetic energy of nucleus, crystal as whole body or is transformed into lattice vibrations (phonons). Due to this fact, the emission and absorption energy spectra may be different and without any overlaps, which prevents the resonance effects from happening.

In the case of free, stationary nuclei, momentum and energy transfers are so high, that the emission and absorption spectra are shifted to different directions by large energetic values, which makes the resonance absorption impossible to observe. However, the nucleus bounded into the crystal lattice is a different case - the crystal as whole body will absorb the momentum. If we consider, that the entire crystal has much larger mass than the nucleus, the energy transfer into crystal's kinetic energy will be very small - the gamma photon energy remains nearly unaltered. Thus this can

be considered as recoilless emission/absorption and the energy spectra have overlap, which makes the resonance absorption (Mössbauer effect) observable.

It also worth mentioning, that there is also a third case - the free nuclei in thermal motion. The velocity of nuclei is guided by Maxwell's statistics and the spectra become widened by Gaussian shape. At higher temperatures, this effect may result into spectra overlap and makes the resonance absorption observable. However, this technique is not much developed yet and has only a small field of application.

5.1.2 Interaction of nuclei with local fields

The nucleus bound inside the lattice surrounded by electrons arranged according to the chemical bonds has perturbed energy levels, which is due to the interaction of nucleus with local electric and magnetic fields - what results into the fact, that every phase or chemical constitution has its own nuclear emission/absorption spectrum - Mössbauer spectrum. The quantum physics has very-well known computation techniques to describe these variations in energy spectra - the perturbation theory,

The main properties of nucleus which induces the interactions with local electric and magnetic fields are: atomic number Z , quadrupole momentum Q and its spin I along with its projections. For spectroscopy there are three main interaction:

- Electric monopole interaction - the interaction between the protons of nucleus and the s-electrons (which have non-zero probability of being found inside nucleus). It results into isomer shift of energy levels $\delta = E_M - E_0$. The δ has to be defined with respect to some fixed energy level, for example to the level of the used source.
- Electric quadrupole interaction - the inhomogeneous electric field inside nucleus interacts the quadrupole momentum Q of the nuclei. It results into energy variations with respect to the square of magnetic quantum number $E_Q \sim m^2$ - the states which allows different values of $|m|$ are splitted into sub-states.
- Magnetic dipole interaction - the spin I is tied up with magnetic momentum via relation $\mu = \gamma I$, where γ is a gyromagnetic ratio. This magnetic momentum interacts with magnetic fields inside nucleus and results into nuclear Zeeman effects - the splitting of the states with respect to possible values of magnetic quantum number m . The main difference from the quadrupole interaction is that the spin orientation also matters. The magnetic dipole interaction has a significant role when it comes to study the magnetic properties of materials.

Each of these effects can occur separately or simultaneously with the others.

5.2 Mössbauer spectroscopy

There are several techniques how to obtain the spectra. In laboratories there are dominant the setups employing transmission or backscattering geometry with sample as an absorber and with the doppler modulation of the gamma photons energy. The transmitted photons or other products developed upon deexcitation are detected by appropriate detector with electronics to accumulate the energetic spectra. The energy of gamma photons is varied by the doppler effect - the source is attached to the doppler

modulator (transducer), and by the relative velocity of source and absorber the energy of gamma photons is slightly changed.

For transmission geometry the key concept of measurement is that if we irradiate the sample as an absorber gradually by continuous spectrum of gamma rays with energy around the resonance energy E_0 ("energy scanning" by the doppler modulator), the gamma photons with energy equal to one of the possible transitions (resonant energies) are absorbed with certain probability. Gamma photons with different energies are due to the low cross section of previously discussed matter interactions mostly transmitted. The detector is situated behind the absorber and simultaneously measures the number of gamma counts at defined energy step. The minimal counts are measured around the resonant energy. The transmission geometry Mössbauer spectra can be seen on picture ??.

The concept of backscattering geometry is similar in many ways, but the main difference is the detection of the deexcitation products. The sample's nuclei are excited by appropriate gamma photons and in short time, they decay back into the original state with some effects following - reemission of the "original" gamma photons in random direction, emission of electrons from the shell (ejected by deexcitation energy quanta, also called conversion electrons) followed by characteristic RTG or possibly by auger electrons. The detection of electrons and RTG instead of gamma photons have many advantages and disadvantages and for certain application, the usage of backscattering geometry could be more appropriate. For example, the detection of conversion electrons could handle the in-depth characterization of the sample due to the lesser transmittance of electrons when comparing to gamma photons.

There are also different setups - irradiation by synchrotron radiation (which can produce continuous gamma spectra with high illuminance), using the sample as an source etc.

5.3 ^{57}Fe spectroscopy

One of the isotopes, on which we are capable to observe a Mössbauer effect is ^{57}Fe . As a source is used ^{57}Co , which decays into second excited state of ^{57}Fe by electron capture. The new ^{57}Fe nuclei can deexcite itself by two ways (see fig. 5.1) - by direct deexcitation onto ground state by emitting 136.6 keV photon or by deexcitation onto the first excited state by emitting 122.1 keV photon and after short lifetime emits 14.4 keV photons (or other possible conversion products) when falling into ground state. The ^{57}Fe spectroscopy in transmission geometry is based on detection of the 14.4 keV gamma photons. This thesis is mainly devoted to the application of semiconductor detectors for the detection of these 14.4 keV gamma photons.

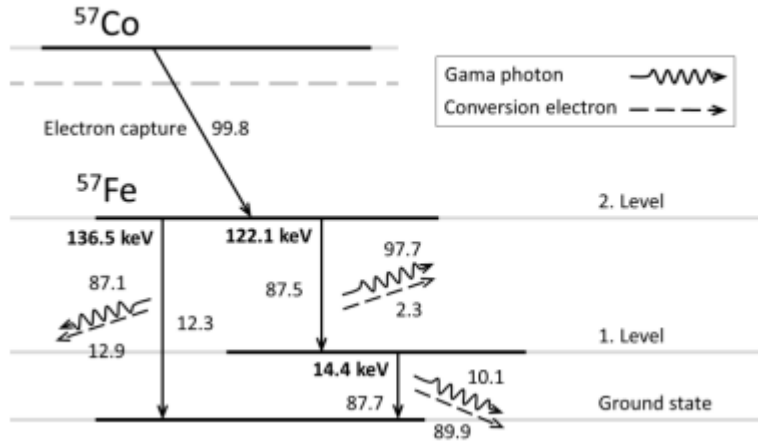


Figure 5.1: Decay scheme of ^{57}Co , taken from [NOVAK2016thesis].

The entire spectrometric setup consist of several parts:

- Source of 14.4 keV gamma - ^{57}Co radioactive nuclei built-in crystal lattice (mostly in a rhodium matrice). The source is attached to the transducer.
- Transducer for doppler modulation. It mostly consists of two coils surrounded by permanent magnets - one for setting the velocity of the source and second for the velocity measurement. The system is driven by PID regulation for precise velocity and energy control. The velocity can be either constant or with constant/varying acceleration.

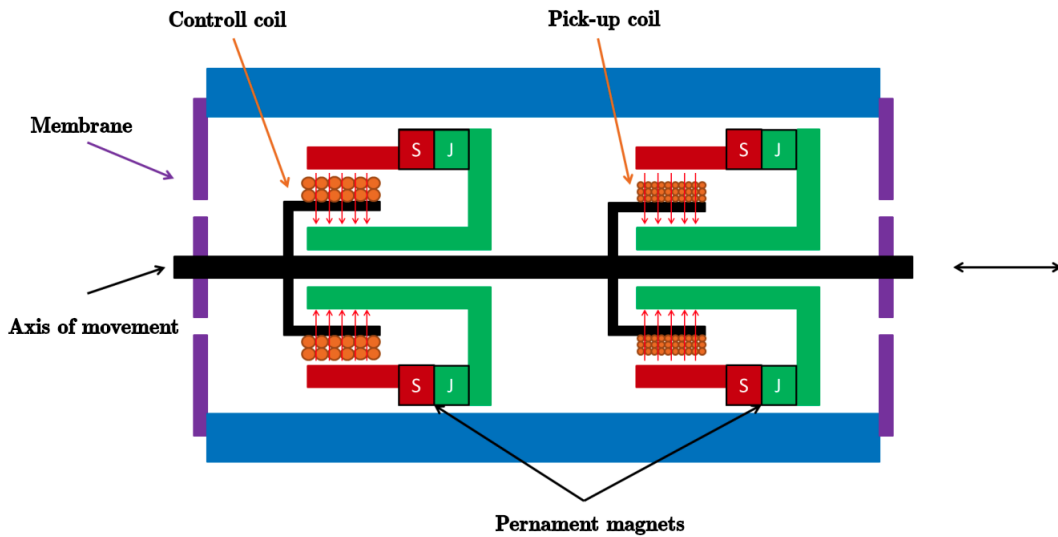


Figure 5.2: Transducer, taken from [STEJSKAL2019thesis].

- detector of transmitted/backscattered gamma radiation, conversion electrons or RTG along with readout and evaluation electronics including amplifiers, SCA's, MCA's etc. It is also necessary to consider, that there are no available detectors with sufficient energy resolution to distinguish energy of perturbed states. Because of that, the count rates are synchronised with the velocity signal (actual

modulated energy), which is used to address the channels for spectra accumulation. The other approach is to address the channels by precise timing.

The functional diagram can be seen on fig. ??.

It is also necessary to consider, that the ^{57}Fe isotope have relative abundance only 2.21% [**compounds**]. Although this fact, the spectra are still measured with very respectable precision and efficiency, which makes the Mössbauer spectroscopy very sensitive measurement method.

5.4 Spectre properties and parameters

Conclusion

The work on thesis was both very hard and experiencing, and thus it can be compared to have to chug a bottle of 50% vodka - get sick, get experienced.