Palacký University Olomouc
Faculty of Science
Joint Laboratory of Optics

MASTER THESIS

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

Author:	**Daniel Staník**
Study program:	N0533A110002 Applied Physics
Field of study:	1702T001 Applied Physics (AFYZ)
Form of study:	Full-time
Supervisor:	Mgr. Aleš Stejskal Ph.D.
Deadline:	April 2024

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 24, 2023

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Daniel Staník

Bibliografická identifikace

Jméno a příjmení autora	Daniel Staník
Název práce	Vývoj spektrometrického řetězce pro detekci nízkoenergetického gama záření s využitím polovodičů.
Typ práce	Diplomová
Pracoviště	Společná laboratoř optiky
Vedoucí práce	Mgr. Aleš Stejskal Ph.D.
Konzultant	Mgr. Leo Schlattauer Ph.D.
Rok obhajoby práce	2024
Abstrakt	Holy moly kihsdlngleiodnglkdngdsrg
Klíčová slova	
Počet stran	22
Počet příloh	1
Jazyk	anglický

Bibliographical identification

Autor's first name and surname	Daniel Staník
Title	Development of a spectrometric chain for the detection of low-energy gamma radiation using semiconductors.
Type of thesis	Master
Department	Joint Laboratory of Optics
Supervisor	Mgr. Aleš Stejskal Ph.D.
Consultant	Mgr. Leo Schlattauer Ph.D.
The year of presentation	2024
Abstract	Holy moly kihsdlngleiodnglkdngdsrg
Keywords	
Number of pages	22
Number of appendices	1
Language	english

Contents

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:

$$\frac{d\sigma}{d\Omega} = \frac{1}{F}\frac{dN_{\mathrm{s}}}{d\Omega} \tag{1.1}$$

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:

$$\sigma = \int \frac{d\sigma}{d\Omega} d\Omega \tag{1.2}$$

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:

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

and integrated:

$$N_{tot} = FAN\sigma\delta x \tag{1.4}$$

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 ($M >> m_e$) 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:

$$I = I_0 exp(-\mu x) \tag{1.5}$$

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:

$$\mu = N\sigma = \sigma(N_a\rho/A) \tag{1.6}$$

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.

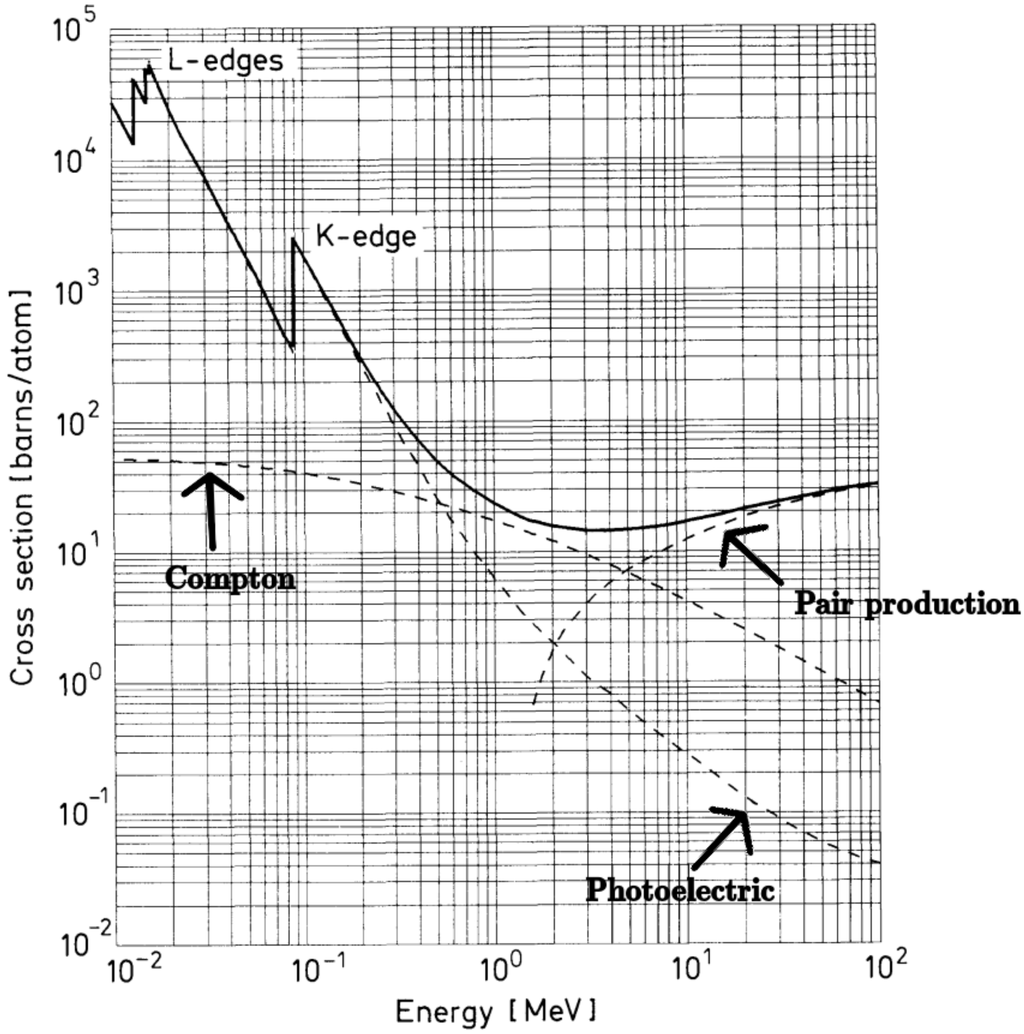

Figure 1.1: Total absorption cross section for high-energy photons. Taken and modified from [**Leo1987-wy**].

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:

$$E = hf - E_b, \tag{1.7}$$

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.

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 imagining - 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 high-energy gamma photons with sufficient energy resolution and efficiency.

3.1 Semiconductor 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. 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 $E_{\mathrm{T}} = kT$, energy in static electric field $E_{\mathrm{s}} = -e\phi$ or as a photon $E_{\mathrm{photon}} = \hbar\omega$. The last one is the most important, because it allows us to use the semiconductors as detectors.

The semiconductor materials have usually form of crystal of diamond structure (two FCC lattices bounded together.).

3.2 Fabrication of semiconductors

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.

3.3 Detection mechanism

If we want to detect a particle or photon, is 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 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 equal to 3.6 eV [**Lutz˙2007**]. Since the Si is an indirect semiconductor, this value is not equal to gap energy $E_\mathrm{g} = 1.12$ 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 interacts with electrons . In case of compton, the scattered photon may escape the detector without any other interaction, which means

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.

3.4 Main noise sources and limitations

3.5 Construction scheme of Instruments for gamma detection based on semiconductors

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

3.5.2 MIMOS 2

3.6 Available semiconductor sensors

To detect Mössbauer 14.4 keV gammas our primary choice was Si due to the high detection efficiency under 25 keV. 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.

3.7 Hamamatsu s14605

3.8 OPF420

3.9 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 Resonace 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 iluminance), 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.

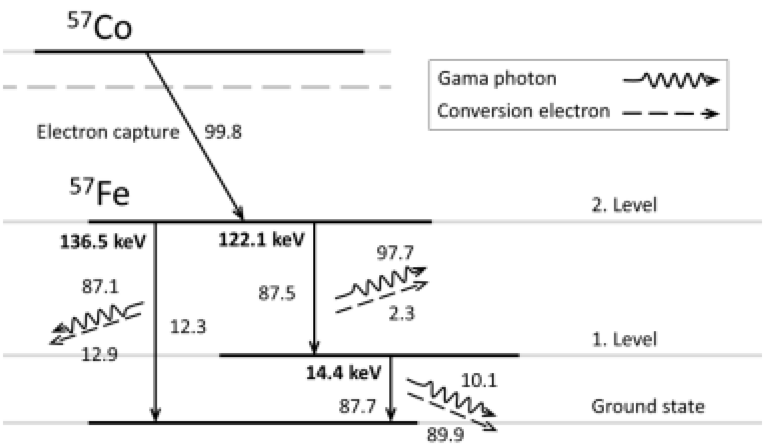

Figure 5.1: Decay scheme of ^{57}Co, taken from [**NOVAK2016thesis**].

The entire spectrometric setup consist of several parts:

- Source of 14.4 keV gamma - ^{57}Co radioctive 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.

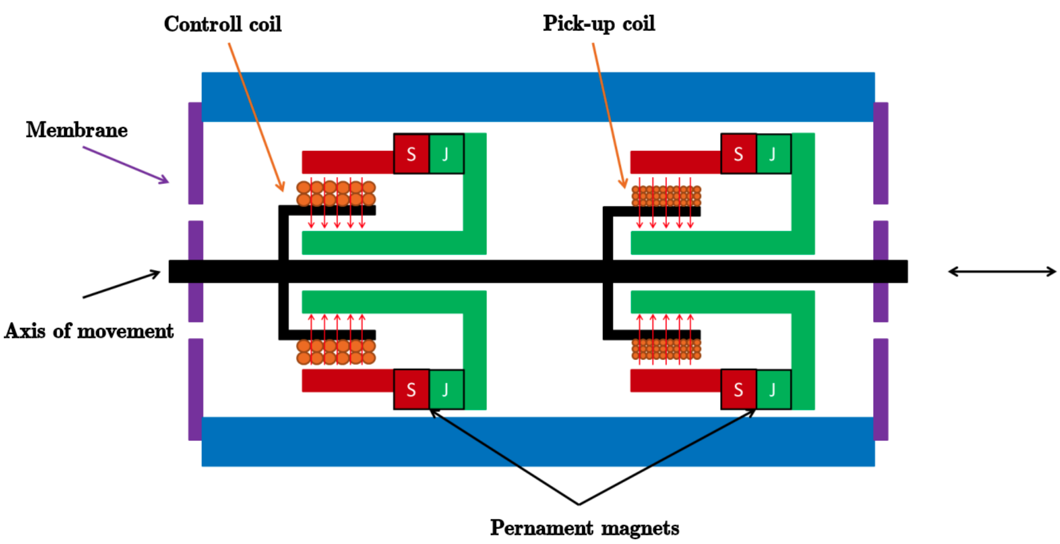

Figure 5.2: Transducer, taken from [**STEJSKAL2019thesis**].

- 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.