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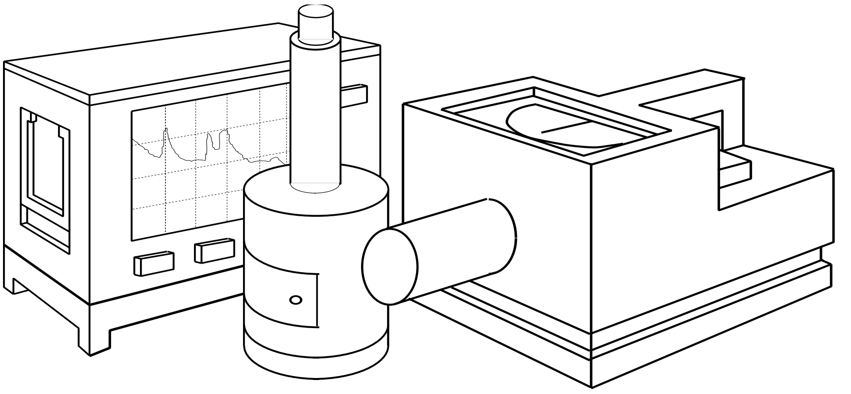
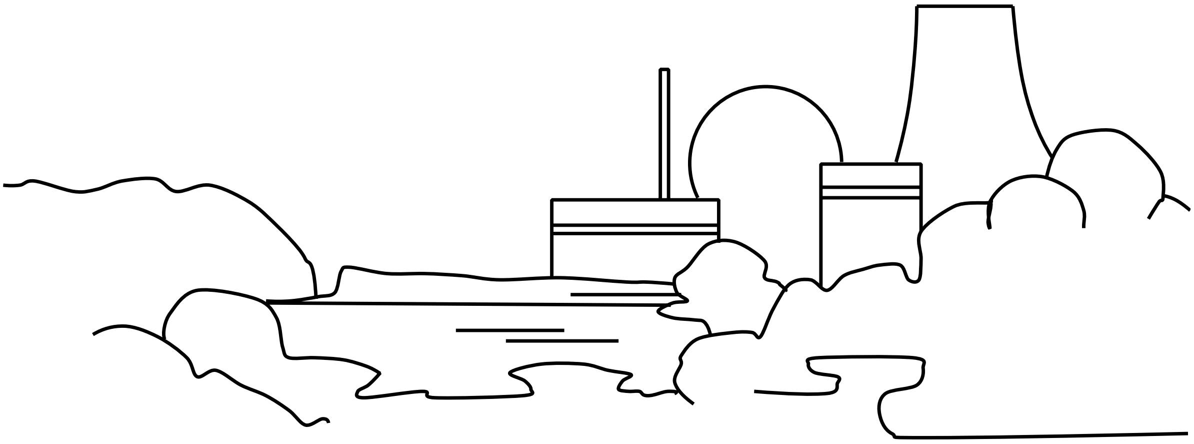


**Reactor Training Course**

**Experiment**

# "Identification of Unknown Radionuclides"
# ( $\gamma$ - Spectrometry )



# Instruction for
# Experiment “Identification of Unknown Radionuclides ( $\gamma$-Spectrometry )"

## Content:

## Figures:

(issued: March 2015)

## MOTIVATION:

Gamma spectrometry is one of the fundamental measuring techniques in nuclear technology for unambiguous, clear identification of radionuclides, even in mixtures of various isotopes.
The experiment is intended to show the basics of the methodology of gamma spectrometry, the properties of most important gamma detectors and how to identify clearly radionuclides. Moreover, the absolute activity of a radionuclide could be determined by means of a gamma spectrometer if it is calibrated absolutely with regards to its efficiency.

## 1. Theoretical Background

### 1.1. Introduction

In nuclear technology, an important task is to identify unknown radioactive nuclides being alone or in a mixture in a sample and to quantify them with regard to their activity or percentage in the mixture. Nowadays, about 270 stable and more than 2000 unstable (radioactive) nuclides are known.

Radioactivity is the transformation of unstable nuclei into stable ones accompanied by a release of energy. Due to this process, the configuration of the atomic nucleus changes. The various transformation processes are accompanied by the emission of ionising radiation.

With respect to radioactivity, a radionuclide is characterised by three properties, which can be used for identification:
- the kind of emitted radiation,
- the energy of emitted radiation, and
- the half-life.

The **kinds of emitted radiation** are:
- alpha radiation (i.e. nuclei of He-4),
- positive beta radiation (i.e. positrons),
- negative beta radiation (i.e. electrons) and/or
- gamma radiation.

Because of the large number of different radionuclides, the unambiguous identification of a nuclide by only four possible kinds of radiation is very limited. In many radioactive decays, not only one kind of radiation is emitted but sometimes several of them in combination. Especially gamma radiation accompanies alpha and beta radiation in almost all cases.

The measurement of the **half-life** is another possibility to identify radionuclides (see also experiment "Activation and Decay of Radioactive Nuclides").

Although the values of half-lives range between parts of a second and $10^9$ years and beyond (i.e. a wide time scale), some nuclides have very similar half-lives and cannot be distinguished within the error tolerances of the experiment. Even more complicated is the case of mixtures of radionuclides with overlaying half-lives. A separation of the particular components would be possible only in a few cases (if the half-lives are very different).

The measurement of the **energy of the emitted radiation**, however, allows an unambiguous identification. In the radioactive decay, the unstable nucleus with its excess of energy gets to the lower energy state of the daughter nucleus by the emission of particles or gamma quants or by resting at an excited immediate state of the daughter nucleus that is left for the ground state only after emission of further radiation. All possible energy levels in an atomic nucleus are predetermined, i.e. through a transformation always the same certain amount of energy is released in form of radiation. As an example Fig. 1 shows the decay scheme of Co-60. The horizontal lines illustrate the energy states of the nucleus. The arrow lines in between mark the kind of decay and its energy. Horizontally shifted lines refer to changes in the charge of the nucleus, i.e. the emission of charged particles. Non-shifted lines mark the emission of gamma quants (no charge). Since energy levels of all existing radionuclides have different values and, consequently, all decay schemes have individual properties, it is possible to identify any nuclide by the measurement of the energy of the emitted radiation. Since almost any radioactive decay is accompanied by gamma emission, spectrometry of this kind of radiation is possible for almost all nuclides. Therefore, gamma spectrometry is the most important and universal method for the identification of radionuclides.

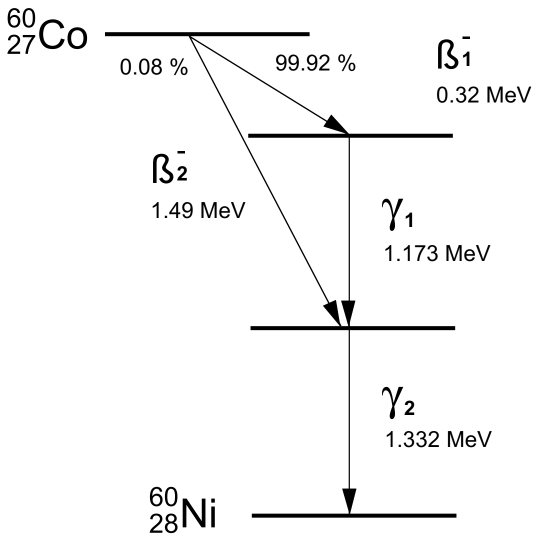


**Fig. 1,** Radioactive decay of Co-60

### 1.2. Interaction of Gamma Radiation with Matter

Gamma radiation is an electromagnetic radiation consisting of gamma quants (= photons). Gamma quants do not carry an electric charge. Therefore, the interaction of gamma radiation with matter differs from that of charged particles with matter. The energy absorption is possible by the **photo effect**, the **Compton effect**, and **pair production**. All these effects cause the production of secondary electrons that are detected (as charged particles) in appropriate measurement devices.

The gamma spectrum is a discrete spectrum. Though, gamma detectors do not only detect distinct sharp lines. This is due to the different possible interactions mentioned above of the gamma radiation with the detector material. Fig. 2 shows a typical measured pulse-height spectrum for a discrete gamma energy (that of the radionuclide Cs-137). At the full photon energy $E_\gamma$, the **photopeak** appears. The gamma quant is completely absorbed by a shell electron of the detector material. In this case, the photon vanishes and a free electron that carries the same energy as the former photon (subtracted by the small ionisation energy of the electron) is released.

If the photon does not lose its entire energy, only an elastic collision with a shell electron of the detector material occurs (**Compton effect**). In this case, the photon transfers only fractions of

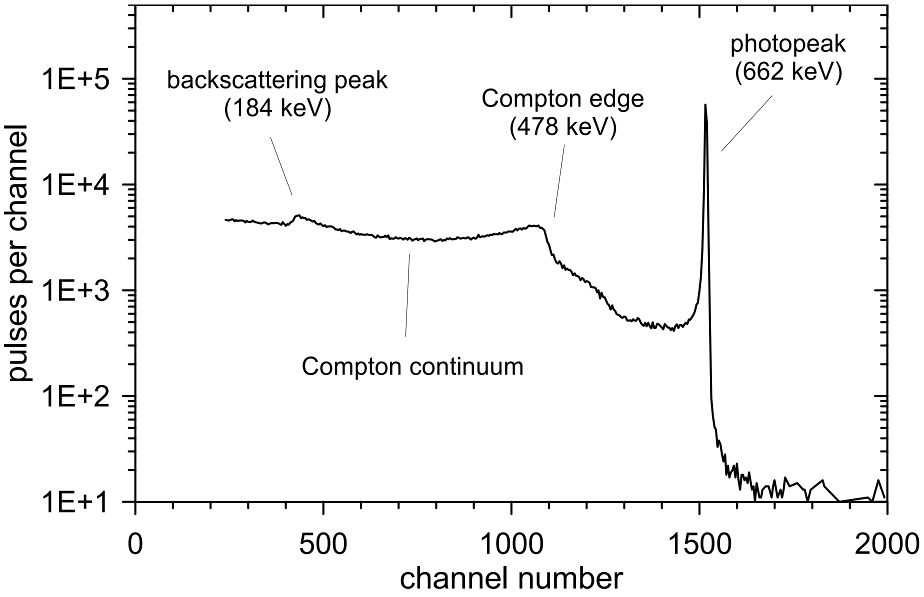


**Fig. 2,** Measured pulse height spectrum of the radionuclide Cs-137

its energy to the shell electron, is scattered carrying still a reduced energy and leaves the detector or can even have a second interaction. The measured pulse height corresponds to the energy of the produced free Compton electron. According to conservation of momentum and energy the maximum energy that can be transferred to an electron is (at an angle of 180°):

$$E_c = \frac{E_\gamma}{\left(1 + \frac{m_0 \cdot c^2}{2 \cdot E_\gamma}\right)} \tag{1}$$

(with $m_0 \cdot c^2 = 511$ keV being the energy equivalent of the electron mass (electron rest energy)). This maximum energy of the Compton effect corresponds to the **Compton edge** in the pulse height spectrum. At lower energies, the spectrum continues with a plateau, the so-called **Compton continuum** that results from energy transmission at angles less than 180°.

At photon energies of some MeV, the full-energy peak is caused by **pair production**. If the energy of the photon exceeds the amount of twice the rest energy of the electron (E > 1.02 MeV), then the Coulomb field of the nucleus can transform a photon into an electron-positron pair. The exceeding energy is almost equally distributed to the electron and the positron as kinetic energy. The pair production is always followed by annihilation of the positron, i.e. after its full slow-down, the positron unifies with an electron of the surroundings under emission of gamma radiation (annihilation radiation).

The ratio of occurrence of the different described effects is determined by the energy of the photon and the material of the detector (Fig. 3). The photo effect is dominating in case of low-energy photon radiation in combination with a high atomic number of the detector material. The Compton effect occurs mainly at intermediate energies at a range of about (0.4 ... 4) MeV.

Photons may also pass the detector without any interaction and re-enter the detector with much reduced energy after being backscattered in the surrounding material. The absorption of backscattered photons corresponds to a **backscattering maximum** at

$$E_{BS} = \frac{E_\gamma}{\left(1 + \frac{2 \cdot E_\gamma}{m_0 \cdot c^2}\right)} \tag{2}$$

that overlaps with the Compton continuum.

In case of a gamma decay with emission of multiple gamma energies or in case of a mixture of various nuclides, the corresponding spectra will overlap. Hence, the identification may be more difficult.

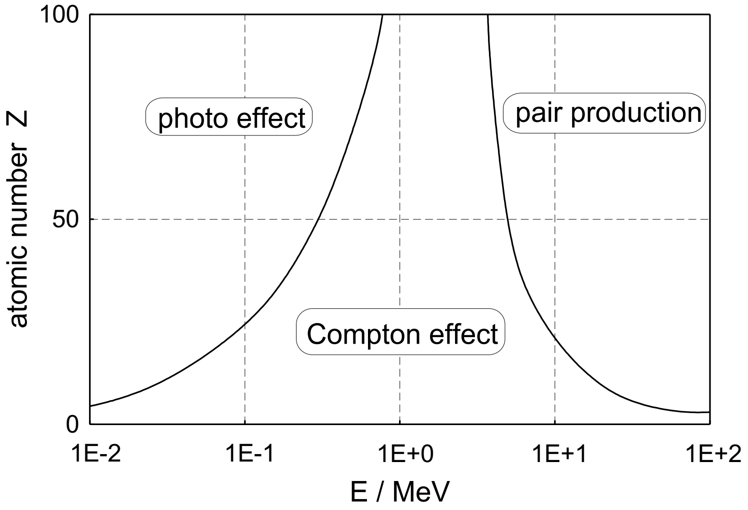


**Fig. 3,** Atomic numbers and energy ranges with dominant occurance of photoeffect, Compton effect and pair production

## 1.3. Measurement of the Radiation Energy

The measurement setup is presented in Fig. 4. The detector is coupled via a preamplifier and a spectroscopic amplifier to a multichannel analyser. In the charge-sensitive preamplifier, the primary charge pulses are converted into voltage pulses (output-voltage is proportional to the input charge). In the spectroscopic amplifier, the pulses are amplified up to a range of 1 ... 10 V. The multichannel analyser recognises the incoming pulses, evaluates them with respect to their height (voltage), which contains the information about the energy, and sorts them into consecutive pulse-height channels. The result is a pulse-height spectrum that contains the energy information of all recognised gamma quants. Each measured gamma quant is a count in the corresponding energy channel of the multichannel analyser. It should be realised that even discrete gamma energies do not cause sharp lines in the spectrum, because of statistical fluctuations of the elementary processes in the detector and in the post-processing electronics. This phenomenon creates from theoretically sharp lines GAUSSIAN bell-curves (peaks). The mean

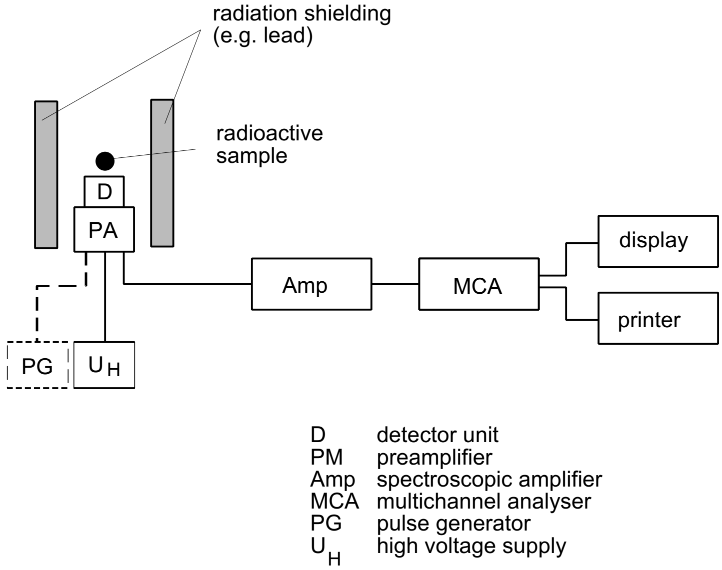


**Fig. 4,** General design of a gamma spectrometer

pulse heights correspond to the energy of the gamma quants. Using a PC programme, the gamma energy spectrum can either be observed on the screen, directly evaluated or saved. The required high voltage is fed to the γ-detector via the preamplifier.

Because temperature effects and voltage fluctuations can shift the electronic amplification of the measurement setup and correspondingly also the channel number of the photopeaks during the measurement, high-quality spectrometers have an automatic amplification control capability. This control uses voltage pulses of an exactly constant pulse height provided by a precision-pulse generator and fed as additional input signal to the preamplifier during the entire measurement. The electronics of the multichannel analyser automatically controls the internal amplification such that the generator peak steadily remains at a constant channel number. The generator pulses are set in such a way that the corresponding peak rises in a region of the pulse-height spectrum where photo lines of the sample are not expected. Usually this is at the very upper end of the pulse-height spectrum.

In most cases, the γ-detectors are surrounded by a radiation shielding, e.g. made from lead, to minimise the influence of the ambient radiation from the environment especially for low gamma activity measurements.

Gamma spectrometry is usually carried out with scintillation detectors (e.g. NaI(Tl)) or semiconductor detectors made from Ge(Li) or more recently made from high purity germanium (HPGe).

**Scintillation detectors** are relatively easy to use and have a high detection efficiency. Special kinds of such scintillators with a tight deep hole in the crystal (where the sample is placed within the detector at the bottom) enables a measurement with almost 4π-geometry.

A disadvantage of scintillators is the limited energy resolution, i.e. the ability to resolve two gamma peaks that are located close to each other as two separate lines.

**Semiconductor detectors** have an excellent energy resolution. Therefore, they are preferably used for demanding analyses in laboratories. Though, the detection efficiency is in general considerably lower compared to scintillation detectors. Using higher efficiencies corresponds to rapidly rising detector costs, even though the efficiency of scintillation detectors cannot be reached. Furthermore, semiconductor detectors have to be cooled (permanently in case of Ge(Li) and at least during usage in case of HPGe) to the temperature of liquid nitrogen (77K). This means further physical and financial efforts.

Fig. 5 shows the pulse-height spectra of the radionuclide Co-60 (gamma energies at 1173 keV and 1332 keV measured with a NaI(Tl)-scintillation detector and a Ge(Li)-semiconductor detector, respectively. The quality of the semiconductor detector proves in form of a clearly better energy resolution of the photo peaks.

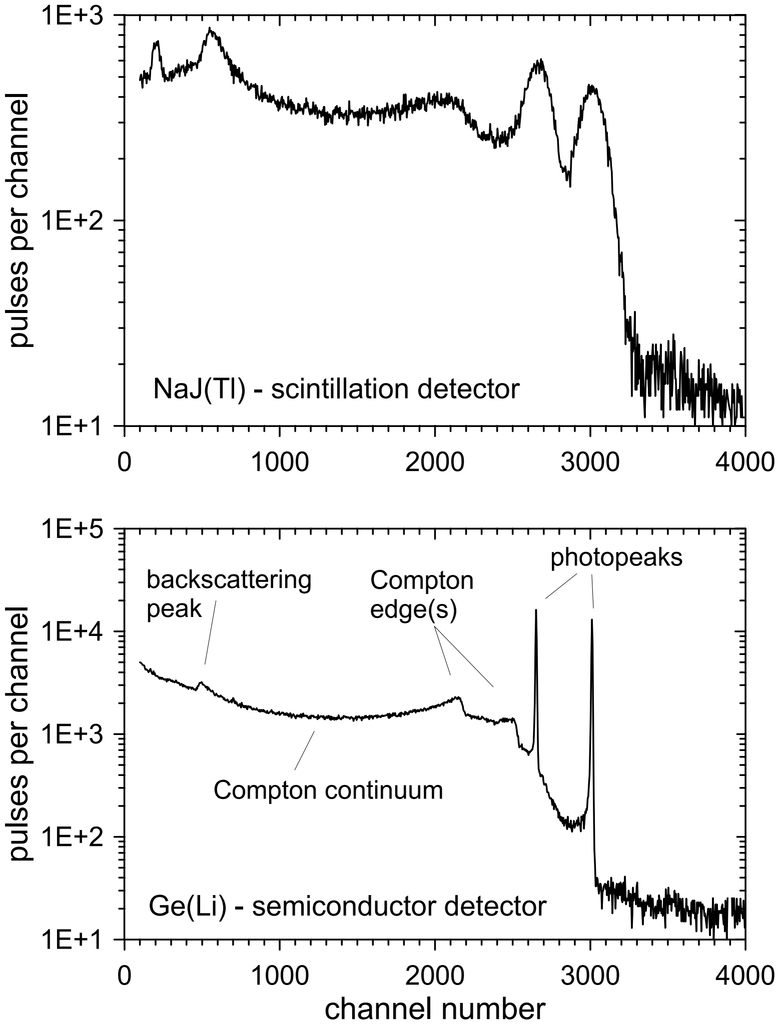


**Fig. 5,** Comparison of gamma pulse height spectra for Co-60, measured by means of a NaJ(Tl) scintillation detector (upper diagram) and a Ge(Li) semiconductor detector (lower diagram)

The resolution (of a detector) is defined as the width of a peak at half of its maximum height (full width at half maximum, FWHM). It is given either as absolute resolution in terms of channel numbers or energy units or as relative resolution if divided by the channel number or energy position, respectively, of the peak maximum.

### 1.3.1. Scintillation Detectors

Scintillation detectors consist of a combination of a luminescent material (scintillator) that is stimulated by ionising radiation to emit flashes (scintillations) and a photomultiplier that converts the flashes into electrical pulses (Fig. 6).

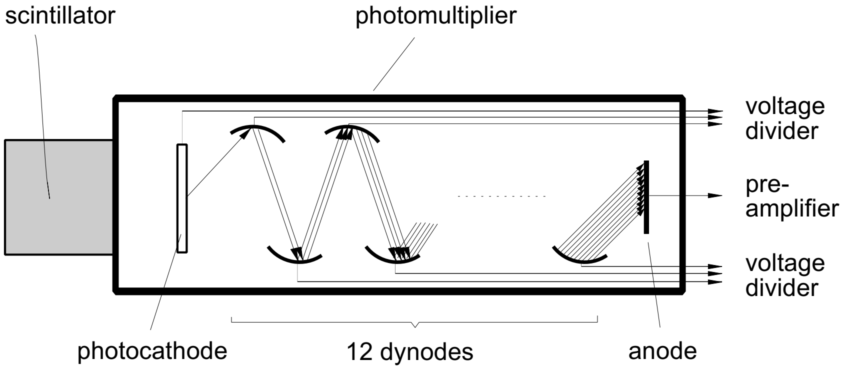


**Fig. 6,** General design of a scintillator-photomultiplier unit

Within the scintillator, the absorbed energy of a gamma quant is converted into light by excitation of the scintillator material and its subsequent return to the ground state. For spectrometric purposes, the absorbed gamma energy and the number of emitted light quants and the subsequent pulse height at the output of the photomultiplier must be proportional to each other.

Inorganic mono-crystals made from sodium iodide (NaI) doped with Thallium for activating the light emission has proved a suitable scintillator material in spectroscopy of gamma radiation (NaI(Tl)-scintillators). Because of their high physical density $\rho = 3.67$ g/cm³, their high content of iodine of 85 wt%, and due to the ordinal number of $Z = 53$, they have a high absorption capability for gamma radiation. Clear crystals that are homogeneously doped and transmissible for their own fluorescent light can be produced in dimensions up to a thickness of e.g. 300 mm or diameter of 400 mm. Also high-energy gamma quants are sufficiently absorbed by these crystals. Commercially available crystals are hermetically sealed (because NaI is hygroscopic) e.g. within aluminium cases, having a glass or plastic window for the light emission. In order to avoid light losses all surfaces of the crystal except for the emission window are not polished but surrounded with a reflecting material, e.g. MgO.

The energy resolution of NaI(Tl)-scintillators is usually given with regard to the gamma line of the radionuclide Cs-137 ($E_\gamma = 662$ keV). Best energy resolution obtained with selected NaI-scintillation crystals and photomultiplier combinations are in the order of about 6 %, common values for commercial scintillator crystals and photomultipliers range between (8...12) % FWHM.

The scintillations from the crystal need to be transmitted to the photocathode of the photomultiplier with only as small as possible losses. Immersion layers (e.g. silicone oil) between scintillator surface and photomultiplier reduce total reflection.

By the photoelectric effect, a few free electrons are emitted on the photocathode. Between the photocathode and the anode of the photomultiplier, a graduated high voltage is applied via a voltage divider and a series of intermediate electrodes (dynodes). In the electric field between each two of the dynodes, the electrons accelerate and liberate further electrons by collision ionisation at the next dynode. This effect leads to a low-noise amplification of the initial number of electrons (factor $10^5...10^9$). Thus, scintillation flashes are transformed into energy-proportional electric current and voltage signals and are amplified in the photomultiplier.

Since ambient light would completely overlap the scintillation effect of the detector and also stimulate the photocathode of the photomultiplier to emit electrons, the entire scintillator-photomultiplier system needs to be light-tightly sealed.

### 1.3.2. Semiconductor Detectors

In the past, mainly scintillation detectors have been used for the determination of gamma spectra. Nowadays, almost exclusively semiconductor detectors are applied for laboratory use. Their main advantage is their high energetic resolution, which is about 10 to 20 times better than that of a scintillator. The energy resolution of a semiconductor detector exceeds that of all other radiation detectors because the energy needed to produce a pair of charge carriers is very low (only about 2.96eV) with the consequence that for a certain amount of absorbed energy a very high number of charged particles with corresponding excellent statistic accuracy is produced. There are the older Ge(Li)-semiconductor detectors and the increasingly used high-purity germanium detectors (HPGe).

**Manufacturing of a Ge(Li)-semiconductor detector:**
In a p-conducting germanium crystal (doped with boron), at 400 °C, lithium atoms diffuse and build up a high Li-concentration at the surface of the crystal. The Li-atoms act as donors, accumulate at inter-lattice positions and form a p-n-junction. Already at room temperature, the inter-lattice atoms are ionised. Consequently, the crystal contains ionised positive lithium atoms. When applying inverse voltage, these ions drift from the n-region to the p-region of the crystal as long as their concentration is higher than that of the acceptors. Due to the formation of boron and lithium ion pairs in the crystal, a high-resistance intrinsic layer (i-layer) is achieved. In this layer, the impurities are eliminated because the negative ions got compensated. Such an arrangement is called pin-structure. The i-layer is an effective barrier with a voltage-independent thickness.

**Mechanism of Ge(Li)-semiconductor detectors:**
High voltage is applied to the contacts (n and p) in inverse direction. As long as no ionising particle penetrates the crystal no electric current arises, since the i-layer has a large electric resistance. If a gamma quant enters the crystal, it generates electron-defect-electron pairs along its trajectory due to photo effect, Compton effect and pair production. These charges are collected at the respective poles by the applied voltage. At the poles, a temporary gain of electric charge, i.e. a potential difference, comes up and causes an electric pulse in the detector circuit (electrical discharge). In a charge-sensitive preamplifier, the charge pulse is converted into a voltage pulse (with an output signal proportional to the input charge).

**Cooling:**
One disadvantage of semiconductor detectors is that at room temperature, even lattice vibrations cause ionisations in the crystal. Therefore, semiconductor detectors need to be cooled during measurements. The cooling is usually made with liquid nitrogen (boiling temperature 77 K or -196 °C, respectively) or recently also by electric cooling units based on the Peltier effect. Ge(Li)-semiconductor detectors need to be cooled even when they are only stored and not in use because at room temperature, lithium atoms are very mobile and would diffuse out of the crystal. Thus, the detector would be distroyed.

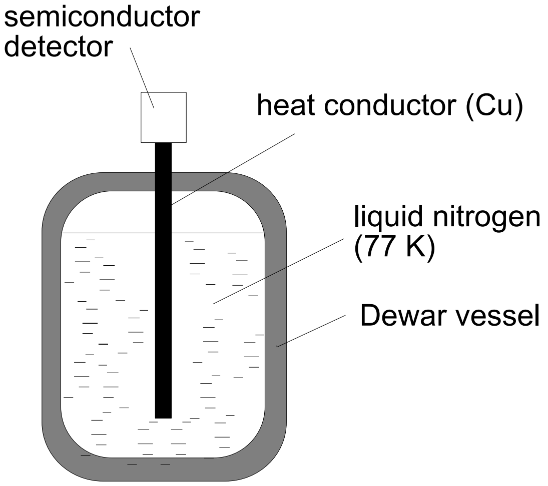


**Fig. 7,** Cooling arrangement of a semiconductor detector

The liquid nitrogen is kept by a thermally well-isolated Dewar vessel (principle of a thermo, see Fig. 7). The semiconductor detector is connected to the nitrogen by a copper finger that removes heat from the detector via heat conduction. To reduce heat losses at the detector, the detector is additionally put into a vacuum vessel. Because heat losses are not entirely avoidable the nitrogen gradually evaporates and needs to be refilled regularly.

### 1.4. Screen Display of the Multichannel Analyser

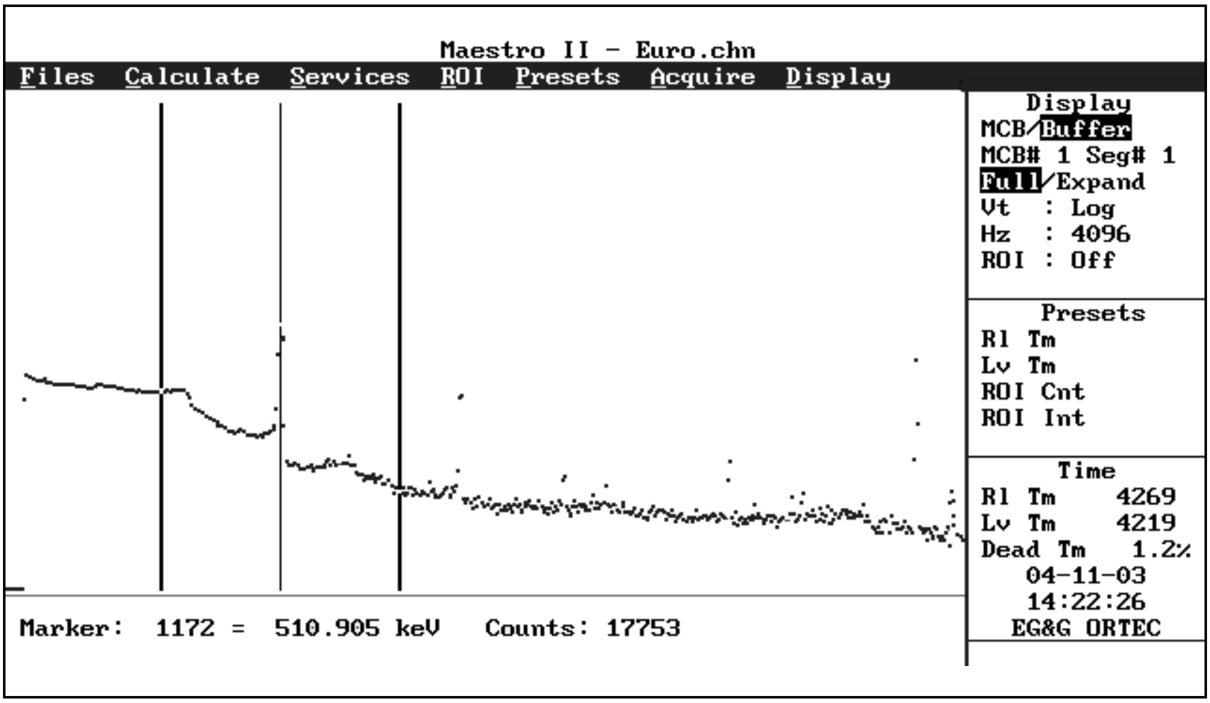
Fig. 8 shows a screen display of the multichannel analyser computer program for an arbitrary spectrum. The vertical cursor in the spectrum marks the selection of a certain channel of the measured spectrum (middle) and the cursor region (left and right). The number of the selected channel is displayed below the spectrum (marker). If the channel axis of the spectrometer has been calibrated with respect to energy, also the corresponding energy of the channel is displayed. Next to this information, the number of counts in the selected channel is displayed.

Important displays within the column on the right hand side of the screen are:

| | | |
|---|---|---|
| Display | MCR/Buffer | Display of the spectrum from the current measurement or from the buffer of the multichannel analyser |
| | Full/Expand | Display of the whole spectrum or only of that part selected in the cursor region |
| | Vt | Scaling of the ordinate axis (vertical) |
| | Hz | Number of displayed channels on the abscissa axis (horizontal) |
| Presets | Rl Tm | Option for preselecting a RealTime for the measurement |
| | Lv Tm | Option for preselecting a LiveTime for the measurement |
| Time | Rl Tm | Elapsed RealTime of the measurement |
| | Lv Tm | Elapsed LiveTime of the measurement |
| | Dead Tm | DeadTime of the multichannel analyser (in %) |

The terms RealTime, LifeTime and DeadTime need to be explained. RealTime is the time period of the measurement according to the clock time. During processing a detector pulse, the multichannel analyser cannot accept additional pulses (it is "busy"). The gate of the analyser is said to be closed. Only after processing the pulse, the gate will be re-opened again to enable the multichannel analyser to accept a next pulse. The time period of an actually open gate is called

LifeTime. Consequently, the DeadTime is that time period with the gate closed. The LifeTime is essential for any absolute measurement of spectra or activity determination. Preferably, the LifeTime should be close to the RealTime, i.e. the DeadTime should be kept small. This can be realised by either fast electronics in the multichannel analyser or by adjusting small count rates.



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Display
MCB/Buffer
MCB# 1 Seg# 1
Full/Expand
Vt  : Log
Hz  : 4096
ROI : Off

Presets
Rl Tm
Lv Tm
ROI Cnt
ROI Int

Time
Rl Tm     4269
Lv Tm     4219
Dead Tm   1.2%
  04-11-03
  14:22:26
  EG&G ORTEC

Marker:  1172 =  510.905 keV   Counts: 17753
```

**Fig. 8,** Monitor screen of a multichannel analyser

## 2. Tasks

### 2.1. Comparison of the Properties of a NaI(Tl)-Scintillation Detector and a Ge(Li)-Semiconductor Detector

The gamma spectra of a Co-60 source, which is characterised by emission of two gamma energies, are measured by both the detectors consecutively.

The setup of the scintillation spectrometer is given in Fig. 9 and that of the semiconductor spectrometer is given in Fig. 10. The parameters of the electronic adjustment are provided by the supervisor. A description of the most important buttons for operating the spectrometer is given in Sec. 3.1.

**Task:** *Place the gamma source Co-60 right onto the respective detector. Discuss the shape of the measured spectra of both the detectors and determine the resolution of the photopeaks. Compare the results of the measurement with the given literature.*

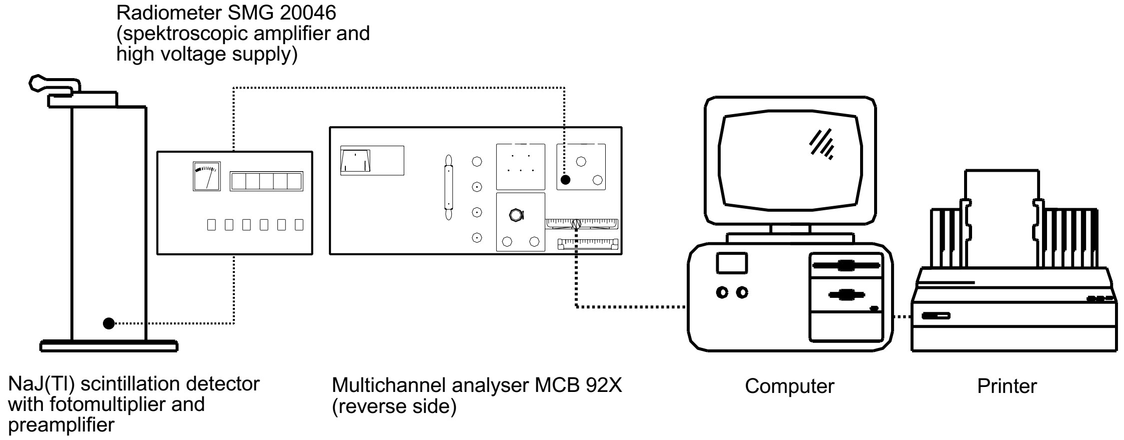

**Fig. 9,** Measuring setup with scintillation detector

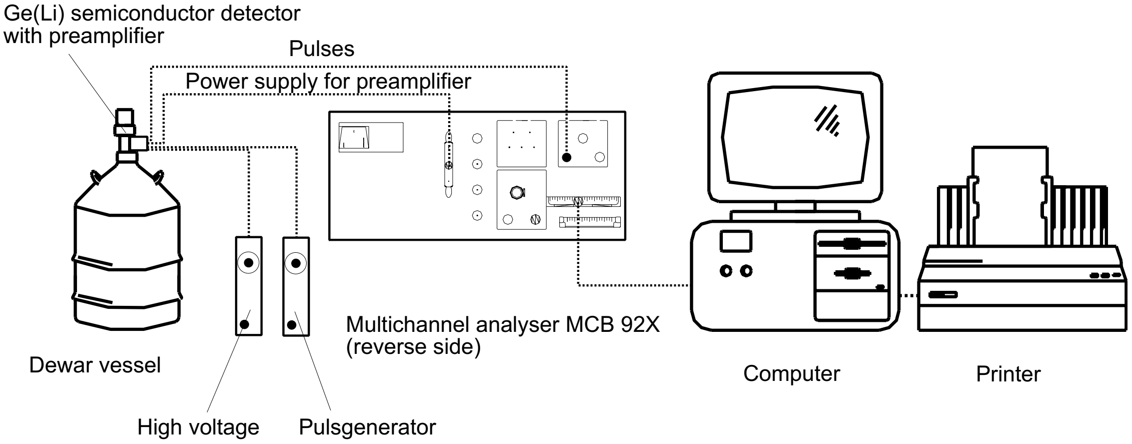

**Fig. 10,** Measuring setup with semiconductor detector

## 2.2. Energy Calibration of the Spectrometer with Ge(Li)-Detector

Analysing spectra of pulse heights, only the channel numbers of peaks and the respective number of pulses per channel are available. In order to link the channel number of the multichannel analyser to the corresponding gamma energy an energy calibration of the spectrometer is necessary.

For the calibration, pulse height spectra of several well known gamma sources (calibration sources) are measured with the multichannel analyser and the channel numbers of the measured photopeaks are related to the well known photopeak gamma energies. A set of such calibration sources is available at the AKR-2 consisting e.g. of nuclides given in Tab. 1.

| Nuclide | Energy [keV] |
|---|---|
| Co-60 | 1173<br>1332 |
| Cs-137 | 662 |
| Kr-85 | 514 |
| Ho-166m<br><br>(main lines) | 184<br>280<br>411<br>712<br>810 |

**Tab. 1,** Energies of the gamma radiation of some calibration sources usable in the experiment

**Task:** *For the given gamma sources, determine the relation between channel numbers and energies of the photopeaks and draw a graphical representation of the function E = f(K). What functional relation can be found?*

It is also possible to let the computer calculate the relation between the channel number and the energy, and subsequently also to display the energy of each channel directly on the screen. Therefore, two points (with the channel number and the respective energy each) taken from the determined function E = f(K) have to be entered in the computer program, which calculates a linear relation. For achieving the best accuracy, the two most-separated point (the lowest and the uppermost) should be used.

The required **keyboard inputs** are as follows:
- Place the cursor onto the channel of the first chosen point (photopeak of the calibration source with the lowest gamma energy)
- Switch to the pull-down menu "Calculate": Alt-C
- Activate "Calibrate": Alt-C
- Type the energy of the chosen channel: ... <CR>
- Repeat the procedure with the second pair
- Type the unit for the energy (keV): ... <CR>

After these steps, the abscissa is calibrated with respect to the gamma energy as can be seen at the screen displaying the respective energy value for each channel number (compare Fig. 8).

**Task:** *Calibrate the spectrometer as explained. Check the calibration with the help of the photopeaks of other calibration sources.*

### 2.3. Analysis of the Time Dependence of the Activities in a Mixture of Isotopes

The element copper comprises the two naturally occurring isotopes Cu-63 and Cu-65 (Tab. 2). When activating copper (e.g. a copper sheet) in a neutron field, the two radioactive isotopes Cu-64 and Cu-66 are being generated. These two isotopes decay with considerably differing half-lives and emit gamma radiation with the energies given in Tab. 2.

Because of the superposition of the two exponential decay curves in the measurement, a simple integral pulse counting does not allow a clear determination of the half-life (see also reactor training course experiment "Activation and Decay of Radioactive Isotopes"). Using gamma spectrometry the measured photopeaks can be related to the respective isotope and their time-dependent intensity can be analysed isotope-specifically.

| Element | Nuclide to be activated | Abundance in element | $\sigma(n_{th},\gamma)$ | Activated nuclide | $T_{1/2}$ | $E\gamma$ |
|---|---|---|---|---|---|---|
| Cu | Cu-63 | 69.1 % | 4.3 b | Cu-64 | 12.8 h | 511 keV and 1348 keV |
| | Cu-65 | 30.9 % | 2.1 b | Cu-66 | 5.1 min | 1039 keV |

**Tab. 2,** Activation data of copper

**Task:** *Activate a copper sample for about 10 min at 2 W reactor power in the tangential experimental channel no. 3 of the AKR. The end of the irradiation time marks the start of the decay time (use a stop watch for the time measurement!). Pre-select an appropriate LifeTime at the gamma spectrometer (see also Sec. 3.1) and measure a few gamma spectra for several decay times each. Relate the measured photopeaks to the two copper isotopes. Determine the net peak areas (that correspond to the intensities) of the photopeaks (see Sec. 3.2). Write the results and also the standard deviations into a table according to Tab. 3 and calculate the ratios of the peak areas of the photopeaks Cu-64/Cu-66. What are conclusions of the measurement?*

| Decay time [ min ] | LiveTime of measurement [ s ] | Net peak areas [pulses] | | Ratio of the peak areas Cu-64 / Cu-66 |
|---|---|---|---|---|
| | | short-lived isotope Cu-66 (5.1 min) $E\gamma = 1039$ keV | long-lived isotope Cu-64 (12.8 h) $E\gamma = 511$ keV | |
| | | | | |
| | | | | |

**Tab. 3,** Protocol of the peak areas of the photopeaks of an activated copper sample

## 2.4. Identification of an Unknown Radionuclide or a Radionuclide Mixture

The nuclide identification of an unknown radioactive sample is the measurement of the gamma energies of the photopeaks and the comparison of the results with the literature. Because all radionuclides have a unique decay scheme (example in Fig. 1) a definite identification is possible. The data of all known radionuclides are available graphically or tabulated in printed or digital form.

In the case that all photopeaks of a measured spectrum can be related to one single nuclide, a pure isotope was measured. If additional photopeaks are identified, these are related to other nuclides and consequently the sample is an isotope mix.

For simple nuclide identification as done in the described reactor training experiment, only the ingredients of the sample have to be determined. An advanced analysis (see Sec. 2.5) allows to calculate the quantitative composition of the sample by analysing additionally the photopeak areas of the spectrum (taking into account the absolute energy-dependent efficiency of the spectrometer, the gamma-emission probability and, if necessary, the activation parameters).

In the training at the AKR, either an unknown sample gets activated in the reactor or an already present long-lived nuclide is measured.

**Task:** *Measure the gamma spectrum of a sample that is provided by the supervisor using the Ge(Li)-semiconductor detector. If necessary, the sample has to be activated in the experimental channels of the AKR before the measurement. The energies of the measured photopeaks have to be recorded in a table according to Tab. 4 (column 1). Using the provided literature data at the experimental place also write both, the gamma energies and the possibly matching nuclides into this table (columns 2 and 3). To account for a slight shift in the energy calibration include also neighbouring gamma energies for comparison, i.e. one keV higher and lower of each measured photopeak. What is the result of your nuclide identification?*

| Measured photopeaks Eγ [keV] | Possibly matching nuclides from the literature | |
|---|---|---|
| | Eγ [keV] | Nuclide(s) |
| | | |
| ⋮ | ⋮ | ⋮ |

**Tab. 4,** Protocol sheet for nuclide identification

## 2.5. Determination of the Absolute Efficiency of the Ge(Li)-Semiconductor Detector and Measurement of the Activity (part of the extended experiment only)

The number of gamma quants of a particular energy emitted by the source differs from the number of detected pulses at this energy in the detector, because

- not all emitted quants penetrate the sensitive volume of the detector (geometry effect),
- not all quants that penetrate the volume of the detector interact within the sensitive volume (detector efficiency effect), and
- not all interactions lead to a complete release of the quantum energy and consequently the resulting pulse does not contribute to the photopeak area of the actual gamma energy (i.e. Compton effect).

The efficiency calibration determines the quantitative relation between the number of gamma quants emitted by the source and the number of pulses being counted in the corresponding photopeak by the detector. The detector efficiency is strongly dependent on the photon energy. At low gamma energies, much more pulses can be registered than at high gamma energies (due to energy dependence of interaction cross sections). Additionally, the efficiency is influenced by the material and the dimensions of the detector, by the geometry of the measurement setup and by the distance between source and detector.

The number of photons that are emitted by the source per time unit is

$$N_{emitted} = A \cdot \gamma \tag{3}$$

with A being the activity of the source (i.e. the number of nuclei that decay per time unit) and γ being the emission probability (or intensity). The emission probability is the fraction of photons which has a certain energy and which is released by decays of the corresponding nuclei. These intensities are provided by the decay scheme of the respective nuclide or appropriate tables.

The number of detected pulses per time unit can be calculated from the measured net peak area and the LiveTime according to

$$N_{detected} = \frac{N}{t_{LT}} \tag{4}$$

The efficiency at a certain gamma energy is the ratio between the numbers of detected and emitted photons:

$$\varepsilon = \frac{N_{detected}}{N_{emitted}} = \frac{N}{t_{LT} \cdot \gamma \cdot A} \tag{5}$$

$\varepsilon$ = efficiency
$N$ = measured net photopeak area
$t_{LT}$ = LiveTime of the measurement
$\gamma$ = photon emission probability
$A$ = activity of the gamma source at the time of the measurement

The detector efficiency at various energies is calculated using this formula for several measurement geometries (e.g. using point sources in a certain distance from the detector or using standardised vessels with a constant volume for liquid samples). If the geometry setup of the measurement is changed, the efficiency calibration needs to be updated.

When plotting the detector efficiency versus the gamma energy with log-log scale, the values get arranged linearly in a wide energy region. Thus, the relation between detector efficiency and gamma energy can be expressed as

$$\ln \varepsilon = a + b \cdot \ln E \qquad \textbf{(6)}$$

For minimising the error of the efficiency calibration, a linear fit should be generated from an appropriate set of pairs of values. This can be achieved by using nuclides with multifold gamma lines or by using a set of several calibration sources. In any case, the absolute activity of the source at the time of calibration needs to be known.

After obtaining the coefficients *a* and *b* in equation (6) by an appropriate calibration, the absolute activity of an unknown sample can be calculated from the measured gamma spectrum by rearrangement of equation (5).

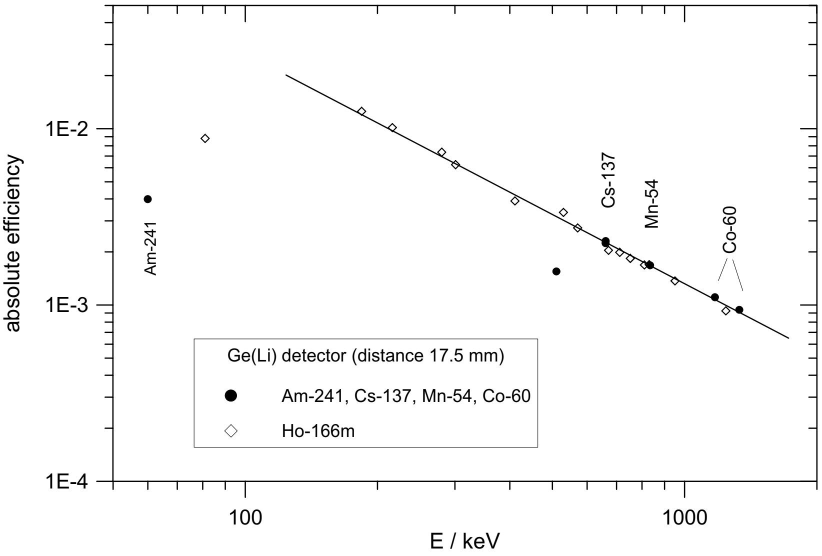

**Fig. 11,** Example of an absolute efficiency calibration of a Ge(Li) semiconductor detector

Examples of calibration sources are given in Tab. 5. The activities of the sources at the time of their production that are needed for the efficiency calibration can be found in the corresponding certificates. These activities need to be converted to the actual activities present at the time of the measurement using the half-lives of the nuclides. An example of an energy calibration is shown in Fig. 11.

| Calibration source | Half-life [a] | Gamma energy [keV] | Intensity [ %] |
|---|---|---|---|
| Am-241 | 432.1 | 60 | 35.90 |
| Cs-137 | 30.17 | 662 | 89.92 |
| Mn-54 | 0.856 | 835 | 99.98 |
| Co-60 | 5.27 | 1173 | 99.90 |
| | | 1332 | 99.98 |
| Ho-166m | 1200 | 81 | 14.00 |
| | | 184 | 100.00 |
| | | 216 | 3.80 |
| | | 280 | 40.79 |
| | | 301 | 4.80 |
| | | 411 | 15.25 |
| | | 530 | 10.00 |
| | | 571 | 6.80 |
| | | 671 | 7.00 |
| | | 712 | 74.48 |
| | | 752 | 16.00 |
| | | 810 | 78.66 |
| | | 831 | 12.00 |
| | | 951 | 3.60 |
| | | 1241 | 1.20 |

**Tab. 5,** Examples of gamma calibration sources (gamma energies and intensities of the lines)

**Task:** *Determine the energy-dependent absolute efficiency of the Ge(Li)-semiconductor detector for a given geometry and present the results in a diagram according to Fig. 11. Calculate the fit function according to equation (6) and give the approximate range of validity with respect to energy. Determine the absolute activity of a given radioactive sample using the result of the efficiency calibration and by rearrangement of equation (5).*

## 3. Operation of the Gamma Spectrometer

### 3.1. Main Control Keys

| | | | Key |
|---|---|---|---|
| **Measurement** | Start | | Alt-1 |
| | Stop | | Alt-2 |
| | Delete | | Alt-3 |
| **Display** | Vertical scaling of the spectrum | | Cursor buttons up/down |
| | View the whole spectrum/cursor region | | F3 |
| | Shift measured spectrum to buffer | | Alt-5 |
| | Switch between measurement display and buffer | | Alt-6 |
| **Cursor** | Cursor movement | slow | Cursor buttons left/right |
| | " | fast | PageUp / PageDown |
| | " | to begin/end of spectrum | Pos1/End |
| | Cursor region | wide | + (NumPad) |
| | " | narrow | - (NumPad) |
| **Save** | Shift spectrum to buffer | | Alt-5 |
| | Pull-down menu "Files" | | Alt-F |
| | Save | | Alt-S |
| | Type in file name (without extension) | | ... |
| | Write a comment (if needed) | | ... |
| **Presets** | Pull-down menu "Presets" | | Alt-P |
| | (e.g.) LiveTime preset | | Alt-L |

### 3.2. Calculation of the Peak Areas of Photopeaks

The area of a peak is the difference between its entire area and the respective background. The background comprises all pulses that do not belong to the photopeak but have been registered by the multichannel analyser in the channels of the peak. The main reasons for the background are:

- natural radiation at the place of measurement (e.g. cosmic radiation, terrestrial radiation),
- artificial radiation at the place of measurement (e.g. operation of the reactor, storage of radiation sources close to the place of measurement),
- background of Compton scattering originating from higher gamma energies of the sample being analysed.

The first two components of the background can be minimised by shielding the gamma detector (e.g. with lead) and/or by storing other gamma sources as far as possible from the place of measurement. If the sample being analysed has more than one gamma line, the third background component is often dominating and cannot be avoided.

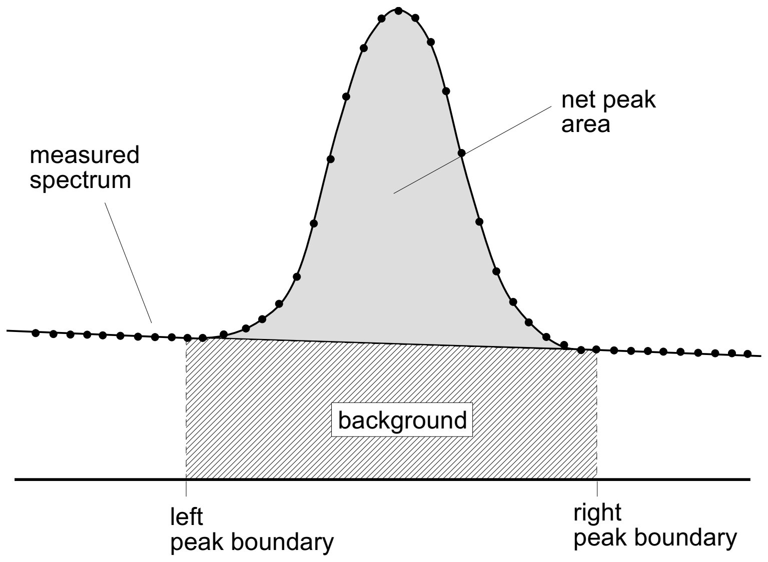

**Fig. 12,** Principle of net peak area determination

For correcting the background, the "ceiling" is assumed to have a linear shape (see Fig. 12). In this case, it can be written

$$B = \left(\frac{C_1 + C_n}{2}\right) \cdot n \tag{7}$$

$$A_g = \sum_{i=1}^{n} C_i \tag{8}$$

$$A_n = A_g - B = \sum_{i=1}^{n} C_i - \left(\frac{C_1 + C_n}{2}\right) \cdot n \tag{9}$$

$B$ = area of the background
$A_g$ = gross peak area
$A_n$ = net peak area
$C_1$ = content of the channel at the left boundary of the peak
$C_i$ = content of the i-th channel
$C_n$ = content of the channel at the right boundary of the peak
$n$ = number of channels between left and right boundaries

The software of the multichannel analyser can calculate automatically the gross and net peak areas as well as the respective standard deviations, which are due to the counting statistics. For these calculations, the following **keyboard commands** are necessary:

| | | |
|---|---|---|
| 1. | Shift the measured spectrum to the buffer | Alt-5 |
| 2. | Display the content of the buffer | Alt-6 |
| 3. | Place the cursor at the left boundary of the peak | |
| 4. | Mark the left boundary of the peak | Alt-R, then Alt-B |
| 5. | Place the cursor at the right boundary of the peak | |
| 6. | Mark the right boundary of the peak | Alt-R, then Alt-E<br>(peak sector is in red color, now) |
| 7. | Move the cursor inside the peak range | |
| 8. | Calculate net peak area | Alt-C, then Alt-A |

The net peak area is calculated by the PC program according to following method. For balancing statistic errors at the lower and upper limits of the chosen peak range, the calculation of the background $B$ averages both, the first and the last 3 channels inside the peak range and uses them for calculating the "ceiling". Hence,

$$B = \left( \sum_{i=1}^{l+2} C_i + \sum_{i=h-2}^{h} C_i \right) \cdot \frac{h-l+1}{6} \tag{10}$$

$$A_b = \sum_{i=l}^{h} C_i \tag{11}$$

$$A_n = A_{ib} - \left( \sum_{i=l}^{l+2} C_i + \sum_{i=h-2}^{h} C_i \right) \cdot \frac{h-l-5}{6} = A_b - B \cdot \frac{h-l-5}{h-l+1} \tag{12}$$

$B$ = area of the background
$l$ = channel number at the left boundary of the ROI (**r**egion **o**f **i**nterest)
$h$ = channel number at the right boundary of the ROI
$C_i$ = content of the i-th channel
$A_b$ = gross peak area
$A_{ib}$ = inner gross area (without both, the first three and the last three channels)
$A_n$ = net peak area
$\sigma_{An}$ = statistical error of the calculated net peak area

# Time-dependent changes in the isotope activities in an activated copper sample

| Decay time [ min ] | Measuring time (LiveTime) [ s ] | Peak areas [pulses] | | Ratios of peak areas Cu-64 / Cu-66 |
|---|---|---|---|---|
| | | short-lived isotope Cu-66 (5.1 min) Eγ = 1039 keV | long-lived isotope Cu-64 (12.8 h) Eγ = 511 keV | |
| | | | | |
| | | | | |
| | | | | |
| | | | | |

Determination of peak areas:

1. Shift measured spectrum into the buffer memory (Alt-5)
2. Show buffer content (Alt-6)
3. Move cursor to left boundary of the peak
4. Fix left boundary of the peak (Alt-R, afterwards Alt-B)
5. Move cursor to right boundary of the peak
6. Fix right boundary of the peak (Alt-R, danach Alt-E)
   (channels in peak region are displayed in red color)
7. Move cursor into the peak region
8. Calculate peak area (Alt-C, danach Alt-A)

**Results:**

# Identification of an unknown activated sample

| Measured γ-peaks Eγ [keV] | Possible nuclides | |
|---|---|---|
| | Eγ [keV] | Nuclides |
| | | |
| | | |
| | | |
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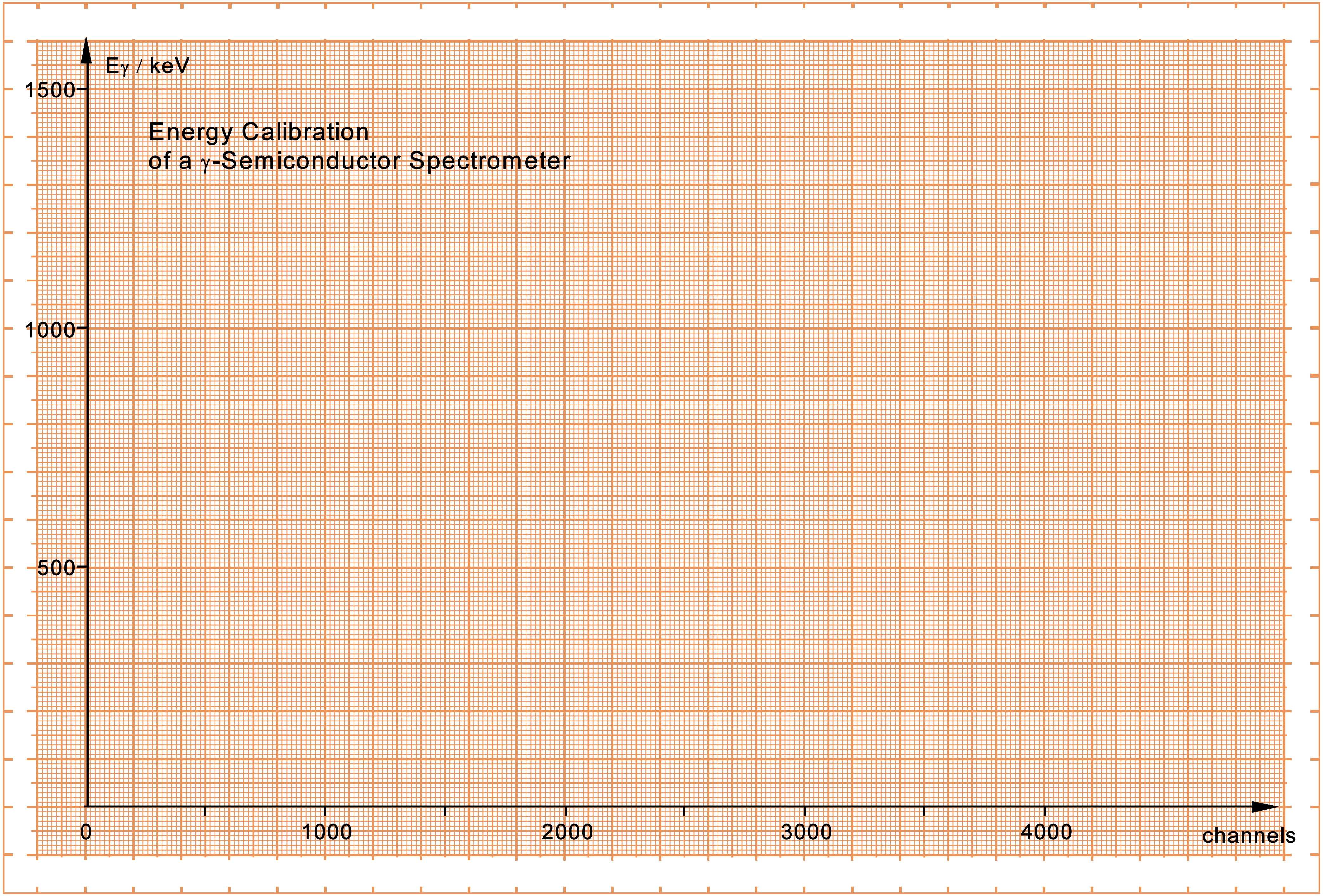
**Results:**


Eγ / keV
1500
1000
500
Energy Calibration
of a γ-Semiconductor Spectrometer
0
1000
2000
3000
4000
channels