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HIGH-PRECISION NEUTRON CAPTURE GAMMA-RAY SPECTROSCOPY USING GERMANIUM DETECTORS IN COMPTON-SUPPRESSION TECHNIQUE

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Abstract

HIGH-PRECISION NEUTRON CAPTURE GAMMA-RAY SPECTROSCOPY USING GERMANIUM DETECTORS IN COMPTON-SUPPRESSION TECHNIQUE. The use of lithium-drifted germanium detectors in Compton-suppression technique will play a very important role in future neutron capture gamma-ray spectroscopy. Over a broad range of energy these devices are superior to all other instruments. High-precision measurements require a favourable peak-to-background ratio and reliable methods in spectrum stabilization, spectrum analysis, calibration and non-linearity correction. The paper reports the procedures that are applied at Karlsruhe and gives some results to demonstrate the efficiency of this technique. Conclusions are drawn on the future potential in neutron capture spectroscopy.

1. INTRODUCTION

Over the last years in gamma-ray spectroscopy increasing attention has been given to the application of lithium-drifted germanium counters since the energy resolution of these devices considerably exceeds that of conventional scintillation detectors. It is understandable that very soon germanium diodes were also being applied to studies of the radiative neutron capture process where spectra with several hundreds of gamma-ray lines have to be analysed and in fact a notable amount of the available data has been accumulated with these instruments. The potential was further increased by operating the detectors in the Compton-suppression mode. In this technique the germanium counter is surrounded by a NaI(Tl), a plastic or a liquid scintillator and Compton events are eliminated to a large extent by means of an anticoincidence circuit. Usually the range of application is between 100 keV and 3 MeV.

In the upper part of this energy range the anti-Compton spectrometer is superior to all other sophisticated instruments both in resolution, precision and sensitivity. For low energies ($\lesssim$ 500 keV) a competitive instrument is provided by the coherent scattering spectrometers which possess uncontested characteristics at very low energies. The boundary in the optimum resolution between crystal diffraction and anti-Compton spectrometer is not fixed. Due to the very different sensitivity, it greatly depends on the cross-section of the material under study. For targets with very high cross-sections ($>$ 1000 b) the boundary may be at energies above 500 keV, while for materials with low cross-sections ($<$ 100 b) the semiconductor system provides the superior instrument over nearly the whole range of application. In this connection it is useful to realize that about

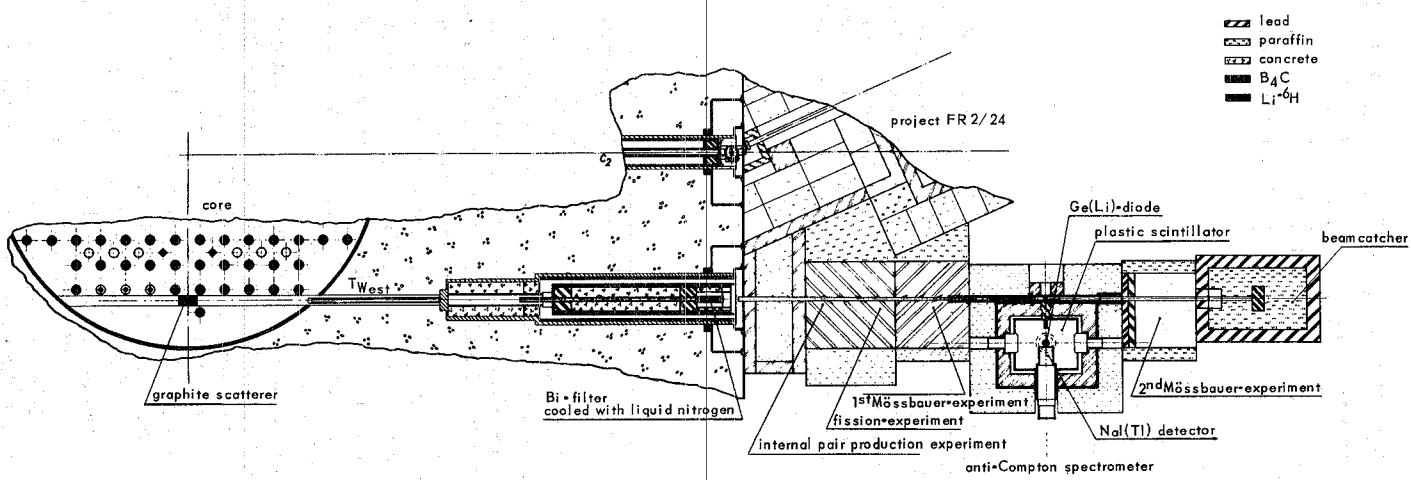

FIG. 1. Schematic diagram of the experimental arrangement at the reactor.

60% of the natural elements have capture cross-sections below 10 b. The correlation between energy resolution expressed in FWHM and precision in energy determination may be quite different for the two devices. In the coherent scattering instrument the energy precision is limited by the optimum resolution in scattering angle, while for the semiconductor spectrometer, in principle, the peak position can be determined to an infinitely small fraction of the line width. Gamma-ray intensities are obtained in a more direct way from semiconductor measurements.

These considerations lead to the conclusion that in spite of the excellent characteristics of the diffraction instruments, the use of germanium detectors in Compton-suppression technique will play a very important role in future neutron capture spectroscopy. However, for obtaining optimum precision with these devices several essential requirements must be fulfilled:

(1) The noise performance should ensure a line width close to the statistical limit; counting-rate effects must be negligible
(2) The peak-to-Compton ratio should be as high as possible or, in other words, the background under the full energy peaks should be low and smooth
(3) The instrument must have a high stability and linearity
(4) The peak positions have to be determined very accurately
(5) The spectra must be carefully calibrated and a suitable non-linearity correction has to be applied to the channel-energy relationship.

The first topic is a general problem in semiconductor spectroscopy and is treated extensively in the literature. Thus we need not discuss it here in detail. The purpose of this presentation is to provide a brief summary of the procedures applied at Karlsruhe on topics (2) to (5) and to draw conclusions from this work on the future potential of anti-Compton devices in neutron capture spectroscopy.

2. PEAK-TO-BACKGROUND RATIO

The anti-Compton instrument, installed in 1966 at the Karlsruhe reactor FR-2, at present consists of an unencapsulated 5-cm^3 Ge(Li) diode, a 50-cm diam. × 40 cm plastic scintillator (NE 102 A) with the germanium counter in its centre, and a 4 in. diam. × 6 in. NaI(Tl) detector placed within a well directly behind the vacuum chamber of the semiconductor diode (for a detailed description see Ref. [1]). The spectrometer is used in external target geometry and is one of five instruments installed at one output of a tangential beam hole which passes through the heavy water of the reflector. A schematic drawing of the arrangement is shown in Fig. 1.

The design of the spectrometer offers the following advantages:

(1) The NaI(Tl) counter ensures a strong absorption of gamma rays scattered in the forward direction. This reduces very effectively the contribution of the ever present intense high-energy radiation to the background under the peaks.

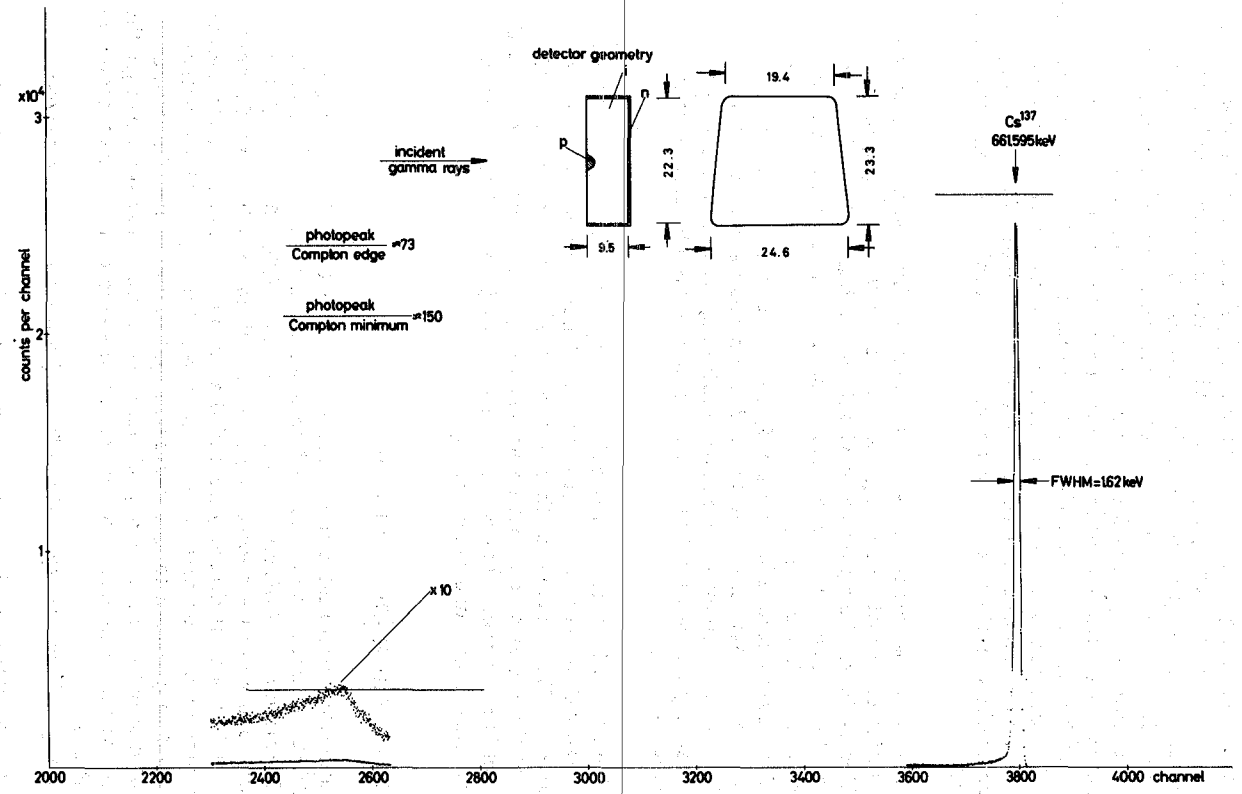

FIG. 2. Sectional display of the pulse-height spectrum of ^{137}Cs taken with the anti-Compton spectrometer.

(2) The plastic scintillator can be used unencapsulated and the reflector, in the form of a water-based emulsion paint, can be kept very thin. Thus the harmful absorption of backscattered soft gamma rays between germanium detector and anti-Compton shield can be minimized. The solid angle for photons escaping out of the anticoincidence shield is very small and is certainly negligible.

(3) The spectrometer is less expensive than other devices which use large NaI(Tl) shields.

A disadvantage of the plastic scintillator is the lower light output compared to NaI(Tl) and the necessarily larger volume of the detector.

The present performance of the system is illustrated in Fig. 2. For the ^{137}Cs 661.595-keV gamma ray the ratio of photopeak to total height of the background is 73 : 1 at the Compton edge and about 150 : 1 in the minimum of the Compton distribution. The energy resolution including long-term instabilities is 1.62-keV FWHM. In the inset of Fig. 2 the drift geometry of the diode is shown. The absence of a dead layer on the detector front face minimizes the amount of absorbing material for the backscattered soft gamma rays.

The performance of an anti-Compton spectrometer for neutron capture spectroscopy is not only determined by the peak-to-Compton ratio for gamma rays possessing energies within the range being measured (100 keV to 3 MeV), but also by the effectiveness in suppressing the background caused by the high-energy transitions above 3 MeV. Here we have to deal mainly with events from photons scattered in the forward direction and high-energy electrons which deposit only part of their energy in the sensitive volume of the detector. The first component is strongly suppressed by the NaI(Tl) detector, while for reduction of the second component it was found useful to apply a pulse-shape discrimination method to the preamplifier signals of the germanium detector. A detailed description of this circuit has been given elsewhere [2]. To demonstrate the actual performance of the spectrometer a sectional display of a typical capture spectrum is shown in Fig. 3. The spectrum was obtained from a sample of natural erbium (capture cross-section 160 b). Above the energy interval displayed in Fig. 3 more than 300 gamma-ray lines have been detected in the total spectrum. All these lines contribute to the background under the peaks in Fig. 3. The yet excellent peak-to-background ratio clearly illustrates the efficiency of the applied method. Another example is given in the paper on ^{68}Zn presented at this Symposium [3].

Several decisive improvements of the system are still feasible. In single mode the present diode exhibits a peak-to-Compton ratio of approximately 10 : 1 for ^{137}Cs. When the detector was installed, planar diodes were superior to coaxially drifted devices with respect to energy resolution. Thus postulate (1) in section 1 was the dominant criterion. Meanwhile large-volume coaxial detectors with comparable resolving power have appeared on the market. The peak-to-Compton ratio of these devices is considerably higher; values up to 30 : 1 for ^{137}Cs are obtainable.

Other possible improvements concern the detection threshold of the plastic scintillator. The present photomultipliers (EMI 9618B) may be replaced by tubes with photocathodes of higher sensitivity. Furthermore, it would be useful to mount additional tubes on the scintillator, in particular

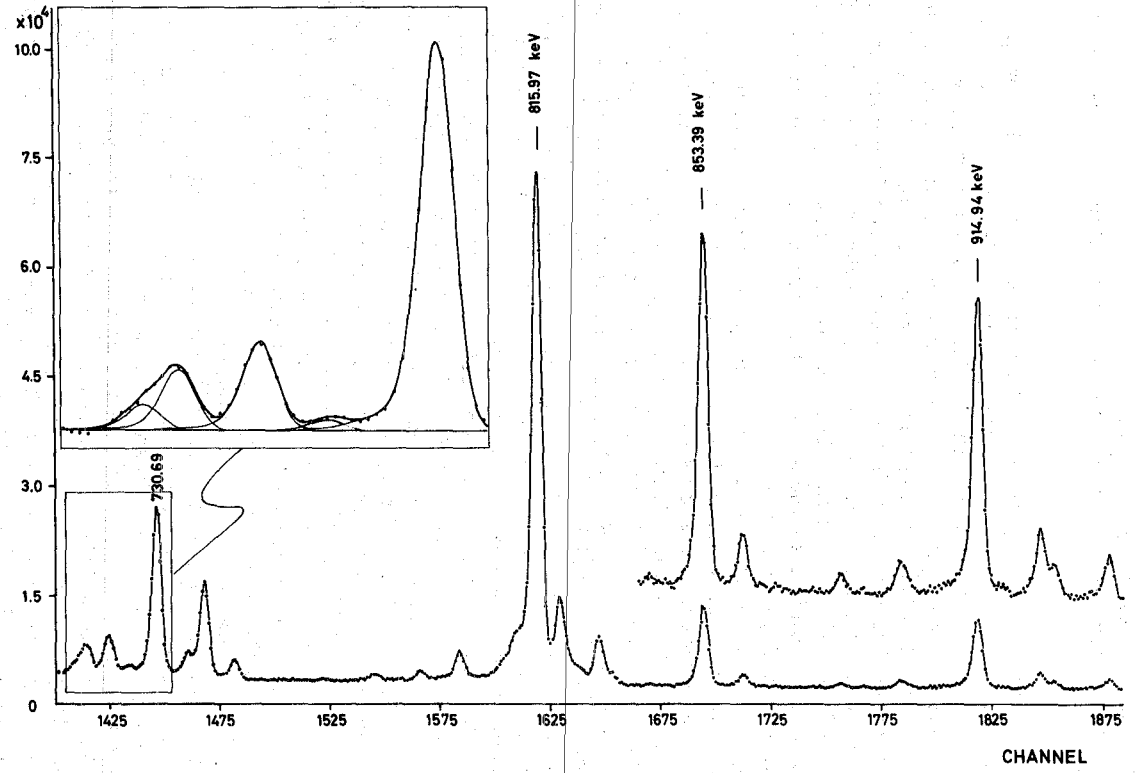

FIG. 3. Sectional display of the capture spectrum from a natural erbium sample.

in the backward direction. Finally, coincidence techniques and cooling of the tubes may improve the signal-to-noise ratio.

We thus can conclude that without sacrifice in energy resolution peak-to-Compton ratios of 250 : 1 for ^{137}Cs are now quite realistic. Suitable modifications of the system will be tackled in the near future.

3. SPECTRUM STABILIZATION

Depending on the cross-section of the sample under study, the measurement of a capture spectrum may cover a period of days or even weeks. Therefore digital stabilization of the amplifier chain and the analog-to-digital converter is a basic requirement for obtaining high precision. This includes both zero drift and gain stabilization. In neutron capture spectra the intensity is distributed over a large number of gamma rays. Thus in most cases no well-isolated natural line with sufficient peak counting rate is available. The presence of an isolated and intense line, however, is very important, if all instabilities independent of their time behaviour are to be eliminated. An alternative procedure is to use an ultra-high precision pulse generator. Here an adequate peak counting rate is attained without producing any additional background outside the full-energy line. Storage of the pulses can easily be prevented by means of an inhibit circuit.

Exceeding demands are made on such a pulser. Since advanced techniques aim at a precision of about 50 eV at 2 MeV (see below), the accuracy of the pulser should be of the order of 10^{-5}. Figure 4 presents an outline of the basic instrument which is now used at Karlsruhe both for gain stabilization and non-linearity correction. The pulser has an instability of < 10 ppm per degC and a non-linearity of $\leq \pm 10$ ppm. The main components of the circuit are a Zener diode network, a Kelvin Varley precision voltage divider, a chopper stabilized operational amplifier for impedance conversion and a mercury relay with pulse shaper. The reference voltage circuit with the carefully aged Zener diode is placed in a temperature-regulated oven and is compensated against variations in temperature and line voltage. Optimum stability is achieved at 65°C where the temperature coefficient of the diode is zero. The impedance matching amplifier ensures the excellent linearity characteristics of the pulser. As regards long-term instabilities this component proved to be the most frequent source of failures[1]. Therefore it is useful to check carefully the warranted performance of the amplifier and to repeat this test at time intervals not longer than a few months. Design and wiring of the pulser output ensure that the influence of the following factors on the output pulse amplitude is kept sufficiently small: the normal amplifier drift rate (1 μV per week), the instability of the pulse shaping capacitor (0.1% per year) and the variation of the mercury relay contact resistance ($\pm$ 1 mΩ). A detailed description of the circuit is given in Refs [4, 5].

If the pulser is only applied for gain stabilization, the impedance matching amplifier can be omitted and the Kelvin Varley voltage divider can be replaced by a simple and less expensive resistance network. For zero-drift stabilization it is convenient to use a natural gamma emitter, e.g. ^{57}Co, with appropriate source strength. The source should not emit any gamma rays within the measuring interval.

[1] "Failure" means in this connection that the pulser output varies by clearly more than 10 ppm.

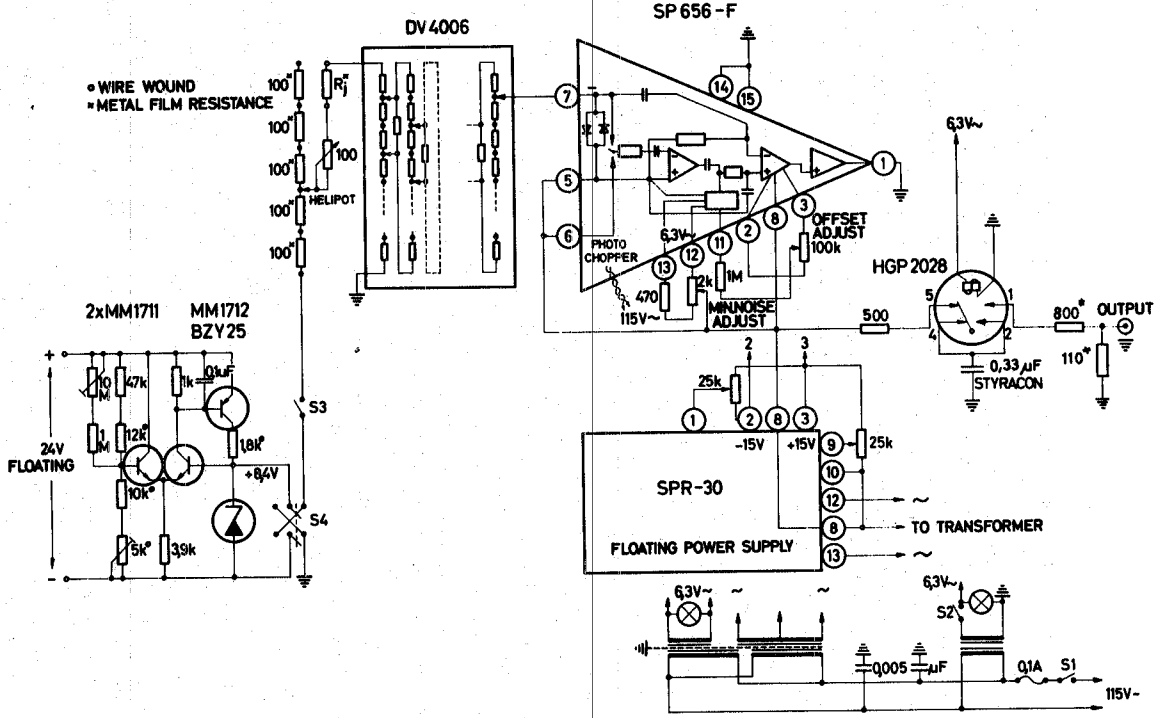

FIG. 4. Outline of the precision pulser.

4. SPECTRUM ANALYSIS

High precision in the energy determination requires utmost accuracy in the evaluation of the peak position. In a computer programme for spectrum analysis reasonable assumptions have to be made on the line shape. An analytical expression should fulfil two requirements: The application to complex spectra must not involve intolerably long computer times and the evaluated curve should fit to the experimental points as well as possible. Very promising results have been obtained by means of the following line shape representation:

$$y = A \exp\{-\lambda (x - x_0)^2\} \quad \text{for } x \geq x_0 - b$$

$$y = A [\exp\{-\lambda (x - x_0)^2\} - B\, x_{cor} \exp(x_{cor})] \quad \text{for } x < x_0 - b$$

where $b = (\lambda^{-1} \ln 2)^{\frac{1}{2}}$ and $x_{cor} = (x - x_0 + b)/b$.

In the case of efficient charge carrier collection in the diode the use of these expressions gives excellent fits for the whole energy range of the spectrometer. This can be seen from Fig. 5 which shows experimental and fitted line shapes for gamma rays at 296, 1173 and 1836 keV. Complex structures are resolved with much success. An example is given in the inset of Fig. 3.

The following procedure is used for the analysis. The spectra are divided into groups each containing at most six gamma lines. Since the background under the peaks is very smooth and only a slowly varying function of energy (cf. Fig. 3), within such an interval the background can be well approximated by a straight line or an exponential function. Energies and intensities are calculated, taking into account a non-linearity correction (section 5) and the response function of the spectrometer. The results are listed by means of a fast printer and each group is plotted in a figure that shows the experimental points together with the fitted background, the fitted gamma lines and the total sum of these curves. Each fit is accomplished with a given number of lines which is chosen to be as low as possible on the basis of a coarse examination of the spectrum. In those cases where the results are insufficient the calculation is repeated, conceding one or more additional lines. This procedure, which guarantees a reliable control by the experimentalist, is clearly preferred to a fully automatic analysis. A detailed description of the programme is given in Ref. [6].

5. CALIBRATION AND NON-LINEARITY CORRECTION

The most suitable gamma sources for energy calibration are the following beta-instable isotopes: ^{57}Co, ^{192}Ir, ^{137}Cs, ^{95}Zr, ^{95}Nb, ^{54}Mn, ^{88}Y, ^{60}Co, ^{22}Na and ^{24}Na. The gamma-ray energies are known very accurately and may be found in Refs [7-10]. Considerable aid in the calibration procedure is provided by the precision pulser described in section 3. A narrow sequence of calibration points can be created with this instrument, even in those energy regions where only few or no natural lines of sufficient accuracy are available.

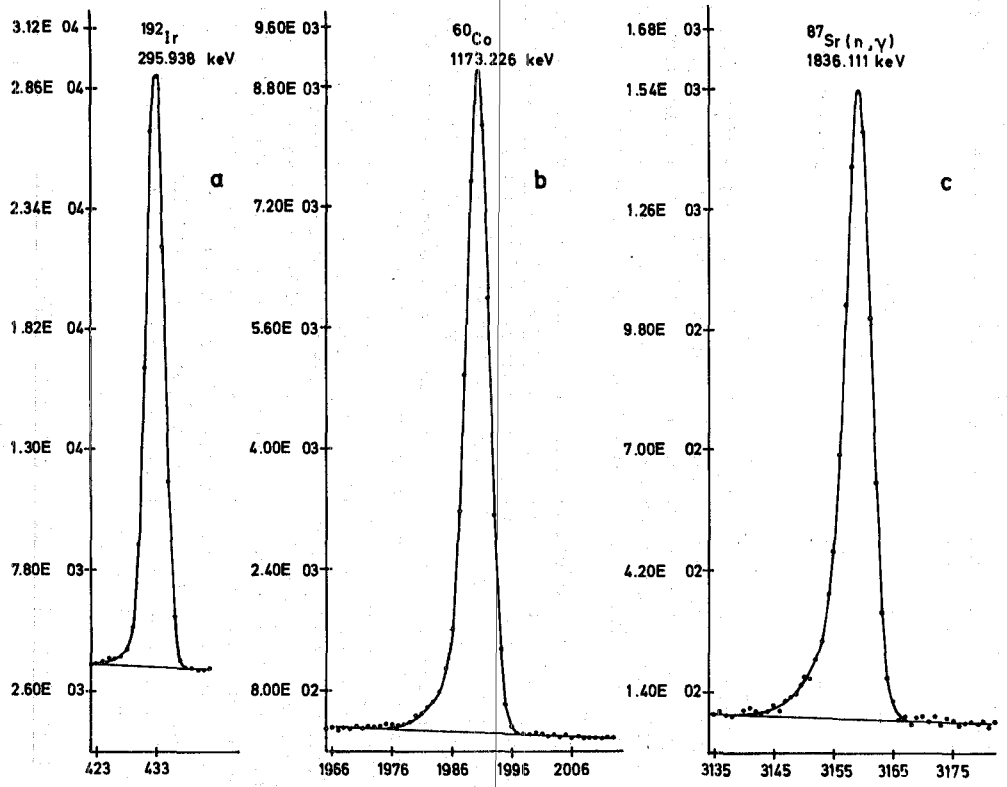

FIG. 5. Experimental and fitted line shape.

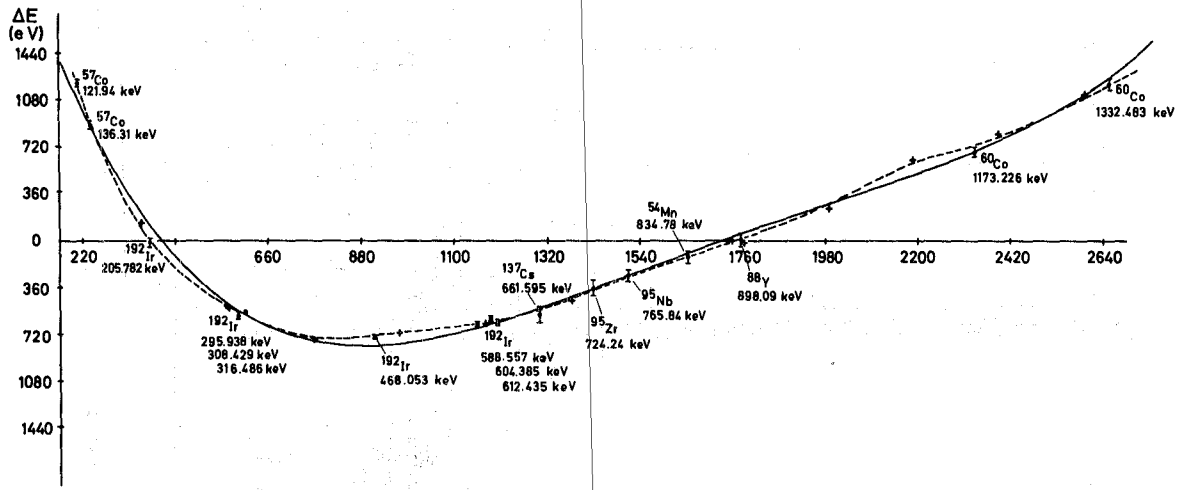

FIG. 6. Typical deviation of the channel-energy relationship from a straight line in eV. The crosses represent calibration points obtained with the precision pulser.

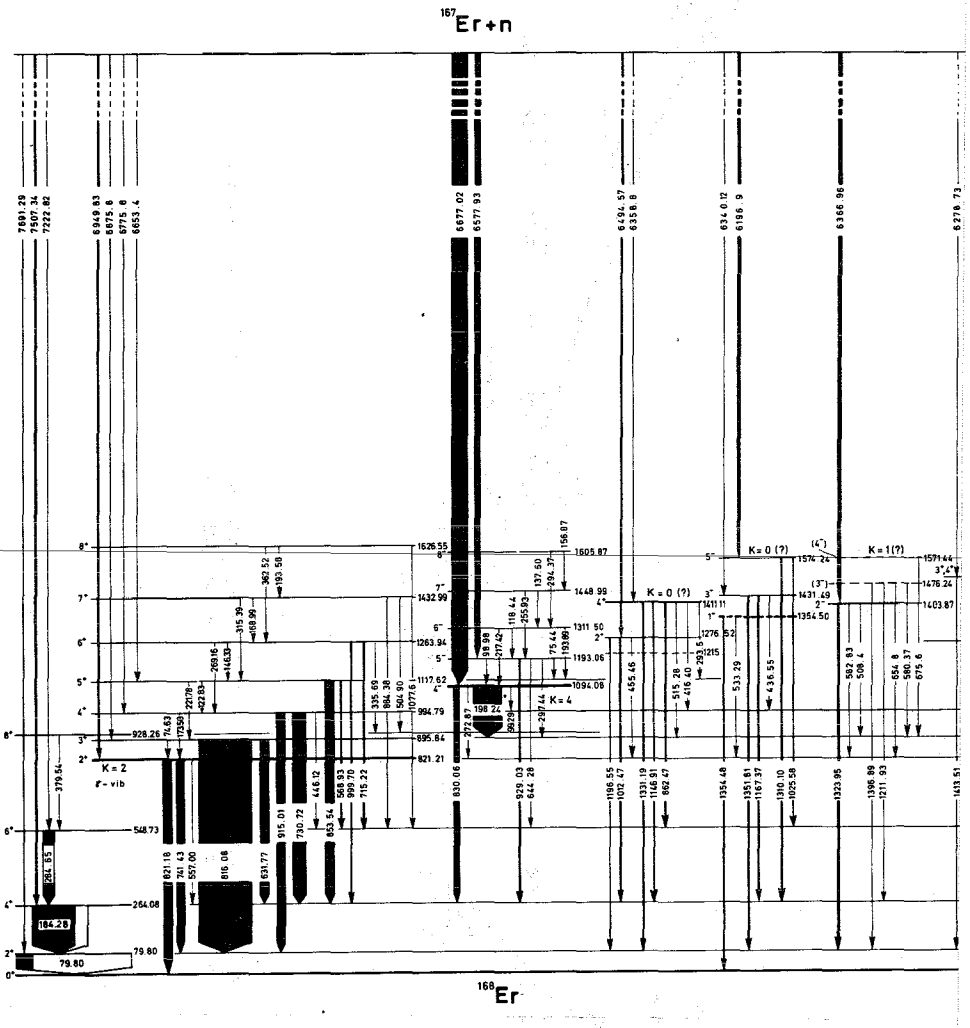

FIG. 7. Transition diagram for ^{168}Er: (a) bandhead energies below 1.5 MeV;

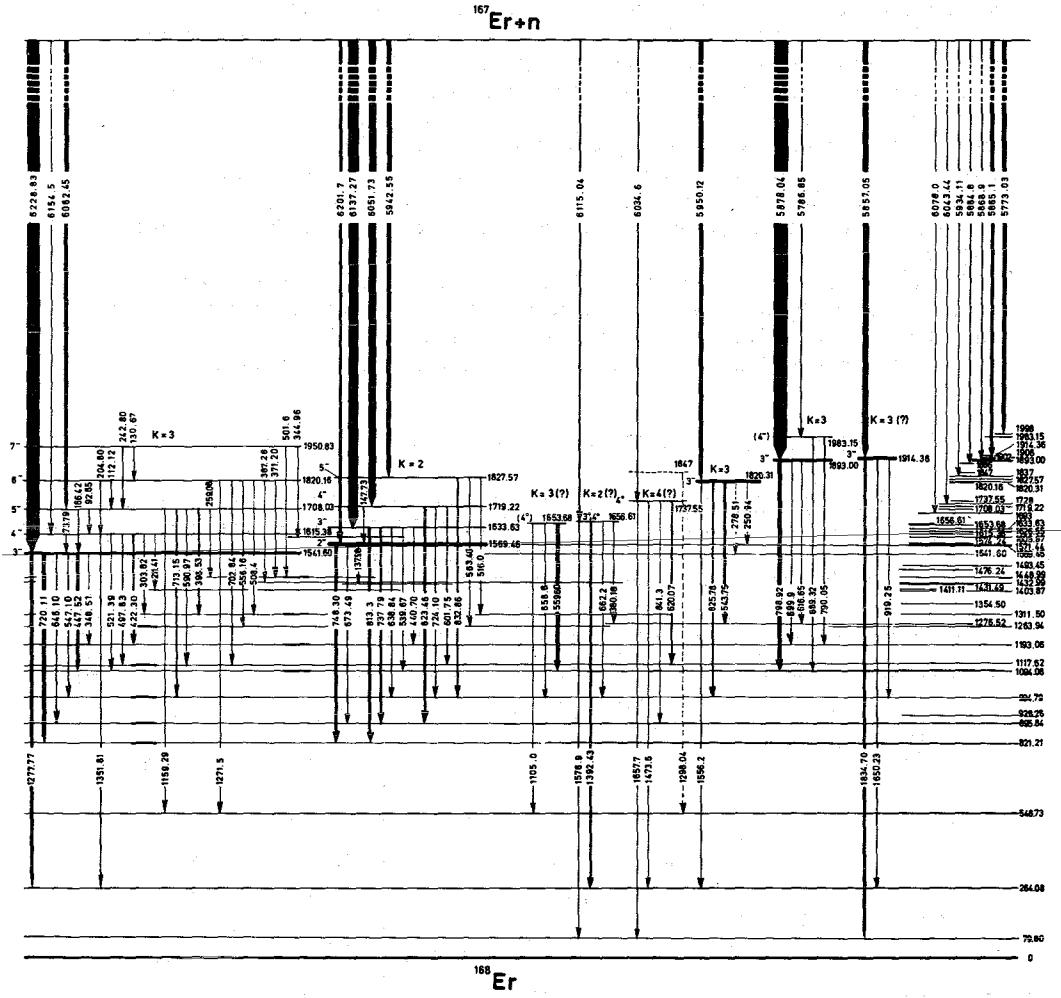

FIG. 7. (b) bandhead energies above 1.5 MeV.

The non-linearity of most commercial amplifiers is of the order of ± 0.05 - 0.1%. Similar values are specified for analog-to-digital converters. Thus for the total system non-linearities of ± 0.1% or more are expected. A typical example for the deviation of the channel-energy relationship from a straight line is shown in Fig. 6. In this measurement the amplifier chain consisted of a cooled FET preamplifier [11] and a main amplifier of type Ortec 410. The pulses were analysed with a 4096-channel ND-160F ADC. The points obtained with the pulser coincide very well with those based on natural gamma lines. The dashed curve represents the 'exact' non-linearity which can be determined with an accuracy of about 50 eV. The full curve shows the result of a fit to the experimental points as obtained with a polynomial of fourth degree. The maximum deviation of this curve from the 'exact' function is 90 eV. In most regions the deviation is much less. We see, however, that a large number of calibration points and a high degree of the fit polynomial are required.

The non-linearity correction discussed here does not account for 'short-range' fluctuations of the differential non-linearity such as the so-called odd-even effect. To minimize the influence of these sources of error, it is convenient to use as small a channel width as possible (≈ 200 eV)

6. EXPERIMENTAL PROCEDURE AND RESULTS

The following procedure is applied for precision measurements of capture spectra: Before the (n,γ) spectrum is taken, a spectrum of the calibration lines is accumulated. The result of this measurement is later compared with that from a similar run performed at the end of the experi-

TABLE I. GAMMA-RAY CASCADES IN ^{68}Zn AND ^{88}Sr

Target nucleus	Cross-section (b)	Gamma-ray energy (keV) E_{γ_1}	E_{γ_2}	E_{γ_3}	Recoil correction (keV)	Level energy (keV)
^{67}Zn	6.9	805.75	1077.35	-	0.014	1883.12
		1883.09	-	-	0.028	1883.12
		670.89	1261.00	1077.35	0.026	3009.27
		1126.07	1883.09	-	0.038	3009.20
^{87}Sr	~17	898.05	1836.08	-	0.025	2734.16
		2733.97	-	-	0.046	2734.02
		1493.41	2733.97	-	0.060	4227.44
		2391.20	1836.08	-	0.055	4227.34

ment. It provides a reliable control that during the experiment no failure (cf. footnote 1) has occurred in the critical electronic components (pre-amplifier, main amplifier, precision pulser, ADC, zero drift and gain stabilizers). In the second run the capture spectrum is taken. After read-out the calibration lines are accumulated on the capture spectrum without memory reset. This ensures that all lines are analysed under the same background conditions. Finally the first run is repeated and the result is used for determining and interpolating the parameters in the line shape representation.

Measurements have been performed on a large number of nuclei. Results on ^{58}Fe (target cross-section 2.5 b), ^{62}Ni (2.0 b), ^{68}Zn (6.9 b), ^{96}Mo (14.5 b), ^{98}Mo (2.2 b) and ^{167}Er (650 b) are briefly discussed in other contributions to this Symposium. Up to 300 gamma rays have been detected in a single spectrum. Because of the heavy shielding of the spectrometer no problems from background lines arise, even for very low target cross-sections. Gamma rays differing in intensity by a factor of 5×10^{-5} have been observed for cross-sections below 10 b. Table I gives an impression on the accuracy that can be obtained for pronounced peaks in the spectra. The absolute errors for the quoted gamma-ray energies are about 50 eV. The errors arising from the fitting procedure and the non-linearity correction are between 5 and 100 eV. The high accuracy permits a reliable application of Ritz' combination principle to excitation energies of several MeV. The accuracy for weak lines is limited by the pure counting statistics. As an example of the efficiency of the method, Fig. 7 presents a considerably extended transition diagram for the nucleus ^{168}Er.

7. CONCLUSIONS

Anti-Compton spectrometers with peak-to-Compton ratios of 250 : 1 for ^{137}Cs are now feasible. By effectively suppressing background events from high-energy gamma rays, a smooth background can be achieved which shows only a slight increase with decreasing energy. The instruments allow energy determinations with an accuracy in the order of 50 eV. For weak lines the precision can be improved by using internal target geometry and thus improved counting statistics. With only a few exceptions precision measurements will be possible over the whole range of cross-sections. Even below 10 b gamma-ray lines that differ in intensity from the most intense peaks by five orders of magnitude should be detectable. Several hundreds of lines can be observed in a single spectrum.

In summary, the application of lithium-drifted germanium detectors in Compton-suppression technique will bring about a considerable improvement in the quality and detail of data that can be attained from neutron capture spectroscopy.

REFERENCES

[1] MICHAELIS, W., KÜPFER, H., Nucl. Instrum. Meth. 56 (1967) 181.
[2] TAMM, U., MICHAELIS, W., COUSSIEU, P., Nucl. Instrum. Meth. 48 (1967) 301.
[3] OTTMAR, H. et al., "Radiative neutron capture studies of nuclear excitations in ^{68}Zn", these Proceedings.

[4] HORSCH, F., MEYER, O., MICHAELIS, W., Rep. KFK 778 (1968); Proc. Symp. Nuclear Electronics, Versailles, 1968.
[5] HORSCH, F., Rep. KFK 558 (1968).
[6] HAASE, V., Rep. KFK 730 (1968).
[7] LEGRAND, J., BOULANGER, J.P., BRETHON, J.P., Nucl. Phys. A107 (1968) 177.
[8] BLACK, W.W., HEATH, R.L., Nucl. Phys. A90 (1967) 650.
[9] BELLICARD, J.B., MOUSSA, A., Compt. Rend. 241 (1955) 1202.
[10] MARION, J.B., Nuclear Data A4 (1968) 301.
[11] TAMM, U., Rep. KFK 509 (1967).