KFK-429

KERNFORSCHUNGSZENTRUM

KARLSRUHE

Juni 1966

KFK 429

Institut für Angewandte Kernphysik

On the Application of Lithium-Drifted Germanium Diodes in Neutron Capture Gamma Ray Spectroscopy

W. Michaelis, H. Schmidt

GESELLSCHAFT FÜR KERNFORSCHUNG M.B.H.

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On the Application of Lithium-Drifted Germanium Diodes in Neutron Capture Gamma Ray Spectroscopy

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W. Michaelis and H. Schmidt

A paper prepared for the IAEA Panel Meeting
"Use of Lithium-Drifted Germanium Gamma Ray
Detectors for Research in Nuclear Physics"
Vienna, Austria
6-10 June 1966

Gesellschaft für Kernforschung m.b.H., Karlsruhe

1. Introduction

Since nuclear reactors provide sufficiently strong neutron sources for investigating radiative neutron capture reactions the study of nuclear structure by observing capture spectra has become more and more attractive. The main physical reason for this trend may be the fact that the information which can be obtained is very sensitive to the specific properties of nuclear excitations. Neutron capture thus reveals important data for a detailed understanding of nuclear structure though the individual excitation energies may be obtained more directly by other nuclear reactions. Because of the complex character of the gamma-ray spectra the earlier work in this field suffered from the inadequate resolution of the available gamma-ray detectors. Considerable progress has been achieved by the development of magnetic Compton and pair spectrometers and most of the data now available in the high-energy region have been collected with these instruments. For analysis of the low-energy spectra up to about 1,5 MeV coherent scattering process instruments with excellent performance have been designed in the past few years. The recent development of lithium-drifted germanium detectors perhaps represents the most significant breakthrough in experimental technique. These detectors can be used over the whole energy range up to 10 MeV. Above 1 MeV their energy resolution is definitely superior to that of all other gamma-ray spectrometers and even in the energy range from 0,6 to 1,0 MeV germanium diodes may compete in resolution with the best bent crystal instruments operating in their optimum conditions. Thus it seems clear that germanium detectors will play an important role in future neutron capture radiation spectroscopy.

Soon after the first realization of such detectors the technique has been applied to capture work in numerous laboratories. At the Karlsruhe research reactor FR 2 experiments in this direction have been performed since May 1965 and several papers have been published or are being prepared for publication *) [1] - [9]. Till now the efforts were mainly directed to the application of the detectors rather than to their technology. The aim of the present status report is (1) to summarize the research being done at Karlsruhe in neutron capture work, (2) to review the capabilities and limitations in the application of germanium diodes and (3) to indicate trends in their future application.

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*) Besides in neutron capture reactions germanium diodes are applied in the spectroscopy of transuranium elements and in fission studies. Examples for the first kind of experiments are described in refs. [10] and [11].

2. Summary of Research and Experimental Methods.

One of the main objectives of neutron capture experiments at Karlsruhe is the study of collective quadrupole vibrations with multiple phonon characteristics in spherical even nuclei. Except for the heavy element region quadrupole surface oscillations are expected to represent the lowest collective excitations. Theoretically the vibrational model predicts the one-phonon state to have $J = 2$ while 2 and 3 surfons couple to a triplet with $J = 0, 2, 4$ and to a quintuplet with $J = 0, 2, 3, 4$ and 6, respectively. The occurrence of such levels is well known from decay studies. However, in decay work the detection of higher-lying levels is limited by the available excitation energy and by the selection rules of beta and electromagnetic transitions. Neutron capture in odd-N nuclei, on the other hand, yields excitation energies of at least 8 MeV and the gamma-ray multiplicity is 3 or more. As a consequence there is a reasonable probability for population of levels having spins several units higher or lower than that of the capture state. Thus the (n,γ)-method seems to be particularly useful for the investigation of multiple phonon states. Further experimental data are very essential for a better understanding of these excitations. At present, the efforts in this direction are focused on the nuclei Fe^{58}, Ni^{62}, Ge^{74} and Sr^{88}. The studies presume a thorough investigation of the total level structure including both collective and shell model states.

Another objective of the Karlsruhe neutron capture experiments involves studies of intrinsic excitations in deformed nuclei. In the mass region $150 < A < 190$ the total beta disintegration energies are such that only low-lying states can be investigated in decay work. Thus similar arguments apply as in the preceding paragraph. The present experimental activity is directed to the odd product nuclei Dy^{165}, Er^{167} and Yb^{169}. These nuclei have the same number of neutrons, but differ in the number of proton pairs. Thus it should be possible to study the influence of the total proton occupation on the neutron level scheme.

In view of the complexity of capture spectra coincidence work represents a very important kind of experiment. Therefore, along with precision measurements of the singles spectra considerable effort has been directed at Karlsruhe to the application of coincidence techniques. High sensitivity of the detectors is essential in this case. As lithium-drifted germanium diodes with sufficiently large volumes have not been available during the last year, the experiments are performed only with NaI(Tl)-crystals. The currently working arrangements are a five-crystal pair spectrometer combined with a single crystal, a triple coincidence set-up and a γ-γ angular correlation apparatus. In any case the precise knowledge of the singles spectra which is obtained from germanium diode measurements is of considerable aid in interpreting the coincidence results since all these experiments suffer from the fact that the resolution of NaI(Tl) detectors is relatively poor. When the assignment of the gamma-ray peaks found in the coincidence spectra or of the gamma rays within the window setting is not obvious, then the following criteria are used for deciding which gamma rays are responsible for the coincidence relationship: (1) the intensity of the lines in the relevant energy interval, (2) the energy difference between the NaI "group" and the nearest gamma lines observed with the germanium detector and (3) the "energy coincidence" of the resulting cascade.

3. The Present Situation in Precision Spectroscopy with Germanium Diodes

In neutron capture radiation spectroscopy with germanium diodes three energy regions may be distinguished according to the different detector capabilities. For each of these energy regions typical examples of capture spectra are given for illustration in Figs. 1 - 3.

Up to about 1.5 MeV interaction of gamma rays by photoelectric effect is the most important process and no difficulties are encountered in assigning the photon energies. Fig. 1 clearly demonstrates the significant advance compared to scintillation detectors. The doublet at 600 keV which corresponds to the two-step cascade de-exciting the first collective states in Ge^{74} is easily resolved. However, with such a simple detector arrangement only limited use can be made of the improved resolution. Usually a few gamma lines with particularly high intensity occur in this energy range giving rise to pronounced Compton distributions which are superimposed on a fairly smooth Compton spectrum from higher-energy transitions. This background makes the detection of weak gamma lines very difficult.

Additional complications arise in the energy range from about 1.5 to 3 MeV where the full-energy peak becomes small and is comparable in intensity to the double-escape peak (Fig. 2). In this region particular care has to be taken as to the interpretation of the spectra. If the structure is complex, very often weak lines cannot be assigned unambiguously.

With simple detector arrangements the most promising results are obtained in the high-energy region above about 3 MeV (Fig. 3, see also Fig. 4). At these energies the diodes are used as double-escape spectrometers. With small detectors ($\lesssim 2$ cm^3) single-escape and full-energy peaks are very small and can be distinguished only in favourable cases. In general, no difficulties arise in analysing the spectra.

As can be seen from Figs. 1 - 3, a considerable reduction of the "background" is essential, if full use is to be made of the detector resolution. Therefore, in addition to aiming at larger detector volumes, three methods are applied at Karlsruhe in order to improve the spectrum shapes.

The first method consists simply of a pulse-shape discrimination on the preamplifier output pulses. A circuit has been developed which is sensitive to the presence of a slow time-constant component in the charge carrier selection [7]. If pulses having such a component are rejected the background under the peaks can be reduced considerably. This is shown in Fig. 4 for the high-energy capture γ-ray spectrum from natural strontium. The detector used for the measurements was a 2 mm deep diode with 2.8 cm^2 sensitive area. The thickness of the lithium diffused layer and of the undepleted germanium was 0.5 mm and 1 mm, respectively. The background is reduced by a factor which increases with decreasing energy. At 7.5 MeV it is approximately 2, at 6.2 MeV a value of 3 is observed. Similar results were obtained with a 5 mm x 4 cm^2 detector. The attenuation of the double-escape peaks is certainly less than 10 %. No effect on the resolution of the diode could be observed. The resulting improvement of the spectra will be particularly desirable when studying weak gamma rays in the presence of intense lines. It is worth mentioning that the method is simple and inexpensive *). The reason for the occurrence of slow time-constant components and for the different effects on background and double-escape peaks is not yet entirely clear. Most probably, the range-energy relation for electrons and the drift of charge carriers from regions with small collecting fields have to be considered.

For spectra involving low-energy gamma rays the effect of the pulse-shape discrimination method is smaller than in the high-energy region. Compton distributions are reduced by 25 - 30 % while the photopeaks remain unaffected. The technique becomes important when studying low-energy gamma rays in the presence of intense high-energy radiation. This is always true of neutron capture spectroscopy.

Another suitable method for background reduction in the low-energy region is the combination of a germanium detector with an efficient anti-Compton shield. Such an arrangement is now being installed at the tangential channel of the Karlsruhe research reactor [9]. The anti-Compton shield consists of a large plastic scintillator with 50 cm diameter by 40 cm length. Behind the solid-state detector a 4"∅x 6" NaI(Tl) crystal is placed in order to effectively reduce the strong forward scattering of high-energy radiation. This design was preferred to a compact NaI(Tl) shield because of its much lower cost. The plastic scintillator is viewed by three photomultipliers with 5" diameter photocathodes. Due to the "shadow" of the NaI(Tl) crystal a length of 40 cm for the plastic scintillator is sufficient. A schematic drawing of the arrangement is shown in Fig. 5. By application of both the anticoincidence and the pulse-shape discrimination method the height of the Compton distribution is expected to be certainly less than 0.5 % of the full-energy peak for the 662 keV Cs^{137} gamma ray. The noise performance aimed at is about 0.5 keV for zero capacitance using a cooled FET preamplifier. First measurements will be made in the near future with a 5 cm^3 diode.

It is important to realize that in addition to the background reduction the double-escape peaks are eliminated. This facilitates the interpretation of complex spectra.

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*) Another pulse-shape discrimination circuit which was developed by Alexander and Goulding [12] for signals from inorganic scintillators is also applicable to solid-state detectors. Tests with the reaction [13] F^{19} (d,nγ) Ne^{20} suggest that similar results may be obtained in neutron capture work.

Above 3 MeV the anti-Compton arrangement becomes ineffective due to the rapidly decreasing photopeak efficiency. Therefore, for precision measurements in the high-energy region the primary detector of the five-crystal pair spectrometer is replaced by a germanium diode. First test experiments indicate that this arrangement represents a very powerful instrument above 3 MeV. If in addition the pulse-shape discrimination method is applied to the solid-state detector the spectra are essentially free from background.

The complex character of neutron capture gamma-ray spectra requires a very high degree of precision. On the other hand, the Ge(Li) detector and the electronic equipment have to be operated in the reactor room where in most cases the conditions for very precise measurements are not ideal. These measurements are more difficult than e.g. decay studies which can be performed under ideal laboratory conditions: (1) long counting periods due to low peak efficiencies, small capture cross sections and/or small amount of isotope under research, (2) environmental conditions concerning instability in temperature and humidity, (3) the unfavourable feature that capture spectra extend over an energy range from a few keV up to 10 MeV and (4) the increased background radiation. Additional precautions have to be taken to overcome these difficulties.

While the background problem can be solved quite satisfactorily with the present techniques [5], [14] long counting periods and temperature instabilities represent a serious problem for precision measurements. Point (3) is connected with this difficulty in so far as in order to obtain optimum results the total counting rate in the electronic equipment is limited to 1000 or 2000 counts per second. Moreover high demands are made on the performance of the biased amplifier. Long term instabilities cause a marked broadening of the line width and the resolution observed in an actual experiment (cf. Figs. 1 - 4) does not reflect the true resolution which is obtained in short counting periods. A typical resolution with the 2 mm x 2.8 cm^2 detector for a run of 13 hours was 9.1 keV FWHM at 7 MeV while the spectrum in Fig. 4 which was accumulated during 48 hours gives a line width of 10.8 keV. In order to avoid this line broadening the data are now taken in shorter intervals, made compatible by means of a special computer program and then summed up. However, the machine program presumes that at least two intense peaks in the short period spectra can be localized with high accuracy. Thus the intervals should be sufficiently long. In order to meet this requirement a temperature controlled box is under development which will receive all temperature sensitive components of the spectrometers except the first loop of the FET preamplifier. This stage will be installed together with the detector in the dewar vessel.

Considering the rapidly increasing expenditure and the presence of instabilities which can not be affected by temperature control it seems to be unreasonable to postulate a temperature constancy in the box of better than ± 1 °C. Hence, there remain some requirements on the instrumental specifications which have to be satisfied.

Our measurements have shown that the main instabilities arise from the ADC and the threshold amplifier the latter one being the most critical element. The following specifications should be aimed at: (1) ADC (4096 channels): long term stability 0.1 channel per 24 hours, temperature drift 0.1 channel per 1 °C; (2) biased amplifier: both gain and threshold stability 0.005 % per 1 °C per 24 hours. As to the ADC none of the older models complies with these specifications. Some very new products seem to meet them to a certain extent at least for 1024 channels. The observed values for commercial biased amplifiers are far worse than the above figures. This is particularly true of the threshold stability.

Another problem in precision measurements is the nonlinearity of the system. Both amplifiers and ADC's currently available show deviations from linearity up to 0.1 or even 0.2 %. Thus at present the only practicable method for obtaining high quality results consists in carefully checking the linearity by means of a very precise pulse generator and in properly correcting the experimental data. This presumes, however, that the linearity relationship is reproducible.

A precision pulse generator is also needed for other reasons. While in the low-energy region the Pb X-rays, the ever present 511 keV annihilation radiation and the full-energy, single escape or double-escape peaks of the photons from the H (n,γ) D reaction can serve as a basis for calibration, there are only a few gamma-ray transitions above 3 MeV which have been measured with sufficient accuracy. The pulse generator should have a performance which is considerably superior to that of the spectrometer system. This requires a linearity and stability better than 0.001 %. A pulser which meets these specifications is presently under development at Karlsruhe.

To take advantage of the higher precision in energy measurements now attainable a computer program is now prepared which allows at reasonable statistics to localize a peak to 1/50 of the line width by a least squares curve fitting. The program uses a modified Gaussian function which takes into account the small low-energy tail of the peaks. It is assumed that the dependence of the line shape on pulse height due to differential nonlinearity of the electronic equipment is negligible.

Some indication of the precision which will be possible when taking the above precautions may be obtained from the results of a recent capture investigation of the Sr^{88} nucleus /5/. The following cascades between capture and ground state could be definitely established by coincidence measurements:

8376 + 897 + 1836 keV = 11 109 keV
8376 + 2734 keV = 11 110 keV
7527 + 850 + 897 + 1836 keV = 11 110 keV
7527 + 850 + 2734 keV = 11 111 keV
6941 + 586 + 850 + 897 + 1836 keV = 11 110 keV
6941 + 586 + 850 + 2734 keV = 11 111 keV

6883 + 1500 + 897 + 1836 keV = 11 116 keV
6883 + 1500 + 2734 keV = 11 117 keV
6883 + 2396 + 1836 keV = 11 115 keV
6658 + 1714 + 897 + 1836 keV = 11 105 keV
6658 + 1714 + 2734 keV = 11 106 keV
6264 + 2113 + 897 + 1836 keV = 11 110 keV
6264 + 2113 + 2734 keV = 11 111 keV
6264 + 3010 + 1836 keV = 11 110 keV

The energy values are those obtained with a germanium diode. Data were taken without temperature regulation and analysis was performed without computer program. The 1024 channel ADC had a long term stability of 0,2 % per 24 hours and was very sensitive to temperature variations. Using a vacuum tube preamplifier the resolution was 6.7 keV FWHM for the photopeak of the Cs^{137} 662 keV gamma ray. As can be seen, the energy sum of nine cascades differs by only ± 2 keV or less from the mean value of 11 111 keV. The absolute uncertainty in the binding energy is less than 4 keV and is mainly due to possible systematic errors in the calibration of the high-energy transitions. The relative deviations when they are >2 keV are attributed to erroneous determination of the peak position in the energy range from 1.5 to 3.0 MeV. It is reasonable to assume that the accuracy can be improved at least by a factor of 5.

In the preceding paragraphs only measurements of the singles spectra have been discussed. However, one of the most important features of germanium diodes is the fact that these detectors are capable of coincidence investigations. Considering the volumes currently available the most promising technique involves the use of a Ge(Li) detector in coincidence with a large NaI(Tl) scintillator. An example for a measurement of this kind is given in Fig. 6 /3/, /6/. The NaI(Tl) counter is used to detect the high-energy transitions proceeding from the capture state. With the availability of larger diodes such coincidence studies will become much easier.

In charged particle spectroscopy with silicon solid-state detectors time measurements by means of a time pickoff circuit have proved to be very useful. An anti-pile up inspector can be easily connected to such a unit. The effect on line width is usually smaller than for double delay line pulse shaping with crossover pickoff technique. When applying the method to gamma-gamma coincidence studies involving a germanium detector difficulties arise with the commercially available circuits as to the threshold for the signal amplitudes (500 to 800 keV depending on detector capacity). Towards lower energies the use is limited by the noise of the fast amplifier input stage. In order to reduce the threshold by about a factor of 10 a fastlow-noise preamplifier was developed /8/. The circuit is a vacuum tube amplifier consisting of two cascode stages and a line driver. The output pulse rise time is less than 4 nsec.

4. Future Trends

The future progress in the application of lithium-drifted germanium diodes to neutron capture gamma ray spectroscopy is closely connected to the availability of diodes with considerably larger volumes. Therefore, in addition to pushing forward the use of the detectors for research in nuclear physics efforts will be directed to the fabrication of large volume diodes. According to a recent paper published by Mann, Janarek and Helenberg [15] it is not unreasonable to aim at volumes in the region of 100 cm^3 or even more. The realization of diodes with these dimensions will permit experiments which cannot at this time be considered. Coincidence measurements will come much more to the fore. The next step involves experiments with two Ge(Li) detectors. While this method will soon be realized in decay studies, it will certainly be restricted for some time in capture work to energy regions with favourable peak efficiencies. A very important goal is the operation of the Ge(Li) pair spectrometer in coincidence with a large volume germanium diode. Another significant application will be the use of germanium detectors in angular correlation studies. The complex structure of capture gamma ray spectra suggests the assumption that the earlier and the present work in this field is affected to a great extent by systematic errors due to the inadequate resolution of the gamma-ray detectors. Approaches to this problem may be made either by replacing one of the scintillation counters or by using a multiple detector arrangement.

Along with the improvement of presently known techniques new physical problems may be attacked when sufficiently large volume detectors are available. This involves e.g. precision studies of gamma ray spectra from neutron capture in the resonance region. Such experiments may be done either by monochromizing neutrons from a reactor by means of Bragg reflection or by applying the time-of-flight method to neutrons from an accelerator. At present studies of low-energy capture spectra, resonance parameters and strength functions are prepared at the Karlsruhe van de Graaff machine and the isochronous cyclotron.

In summary, the application of lithium-drifted germanium diodes will bring about a radical improvement in the quality and detail of data which can be obtained from thermal and resonance capture of neutrons.

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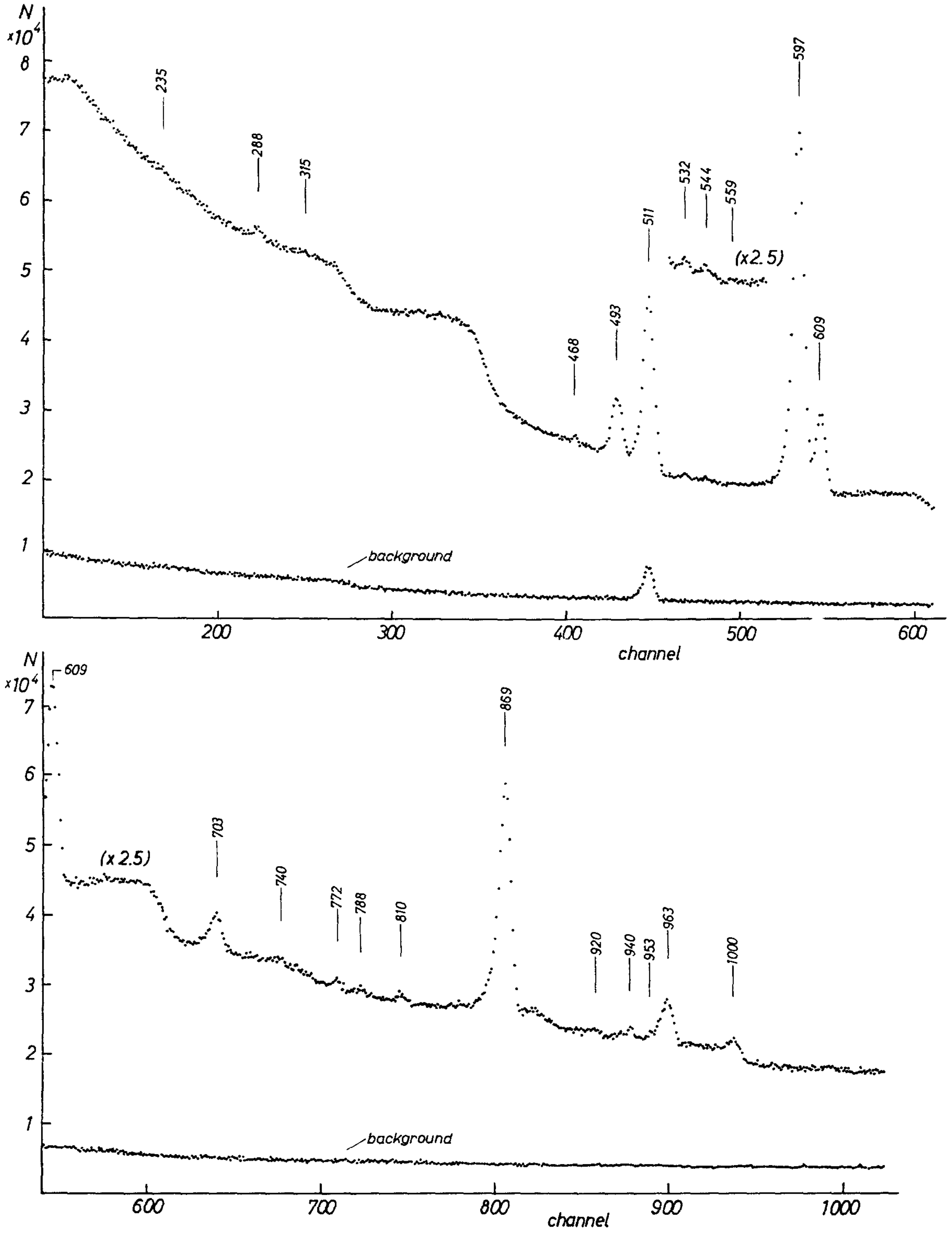

Fig. 1. Singles gamma-ray spectrum from the reaction $Ge^{73}(n,\gamma)$ Ge^{74} taken with a 4.0 cm^2 x 0.5 cm Ge(Li) detector. Energy range from 200 keV to 1100 keV. The target was GeO_2 enriched to 86.1 % Ge^{73}.

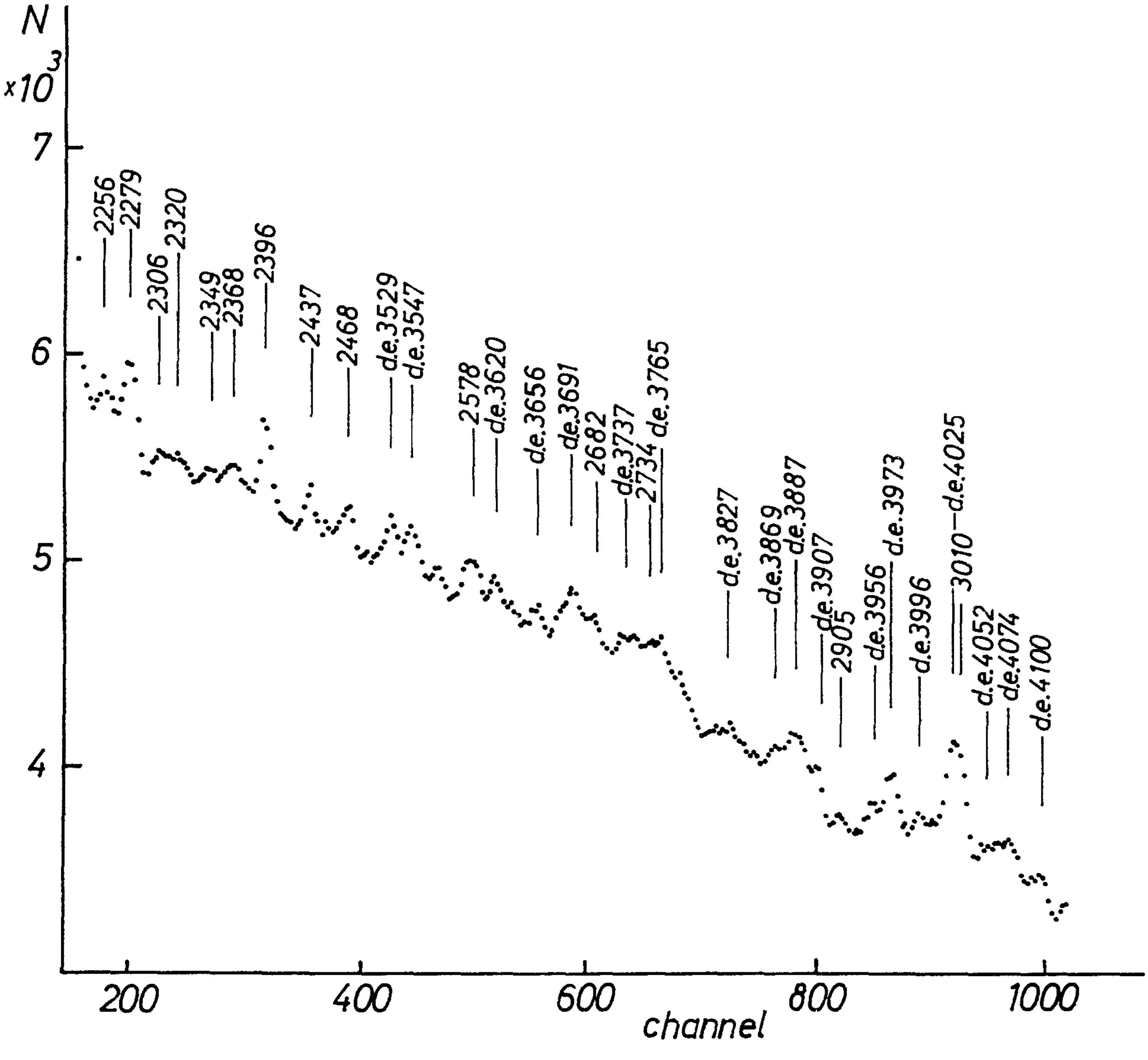

Fig. 2. Singles gamma-ray spectrum from thermal neutron capture in natural strontium carbonate observed with a 4.0 cm^2 x 0,5 cm Ge(Li) detector. Energy range from 2250 keV to 3100 keV. Full-energy peaks are labelled with the gamma-ray energy. Double-escape peaks are labelled d.e.

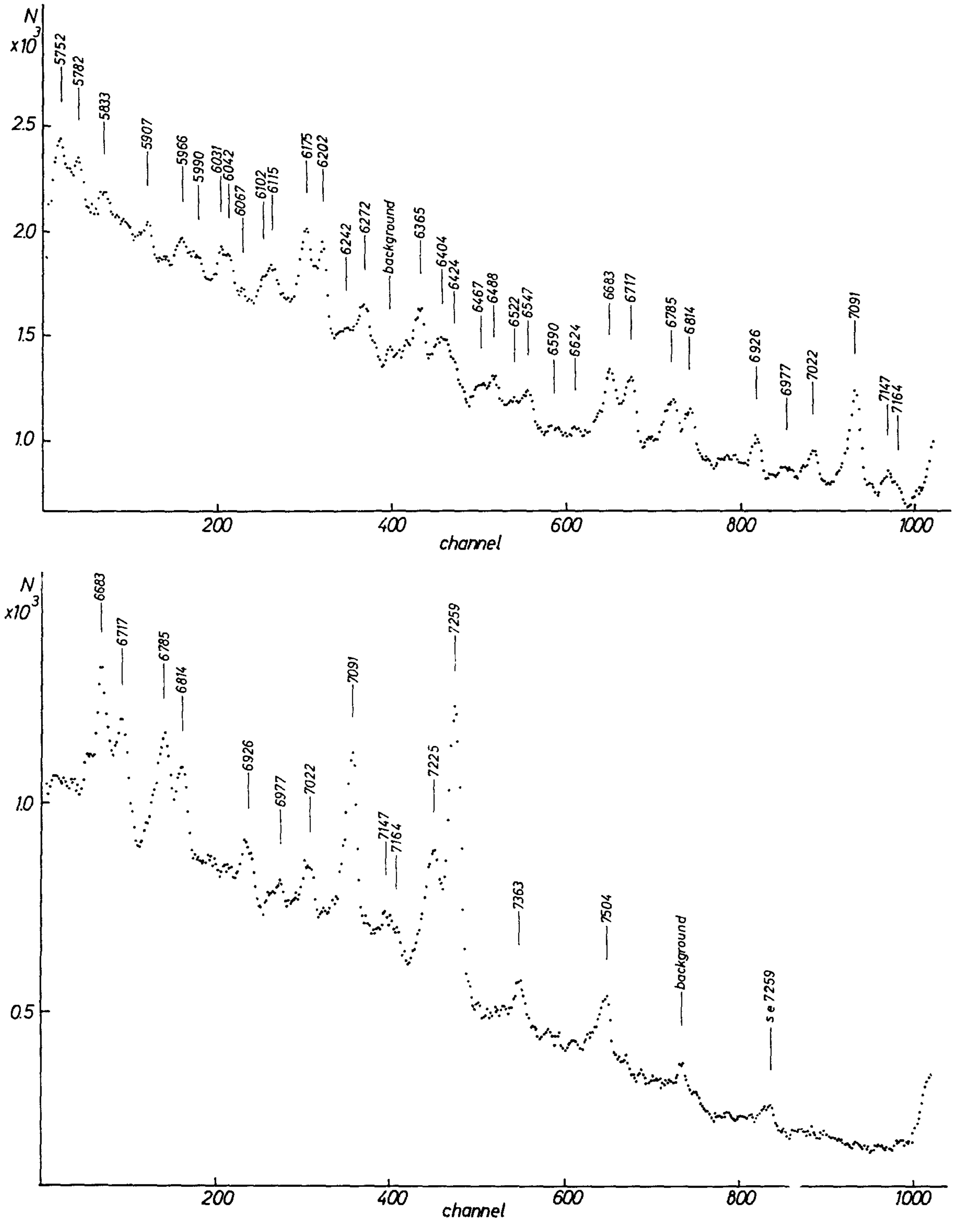

Fig. 3. Singles gamma-ray spectrum from the reaction $Ge^{73}(n,\gamma)\ Ge^{74}$ observed with a 4.0 cm^2 x 0.5 cm Ge(Li) detector. Energy range from 5750 keV to 7900 keV. All peaks except one are assumed to be double escape peaks and are labelled with the gamma ray energy. For the prominent gamma ray at 7259 keV the single escape peak can be distinguished and is labelled s.e. Where background is not smooth peaks are labelled "background".

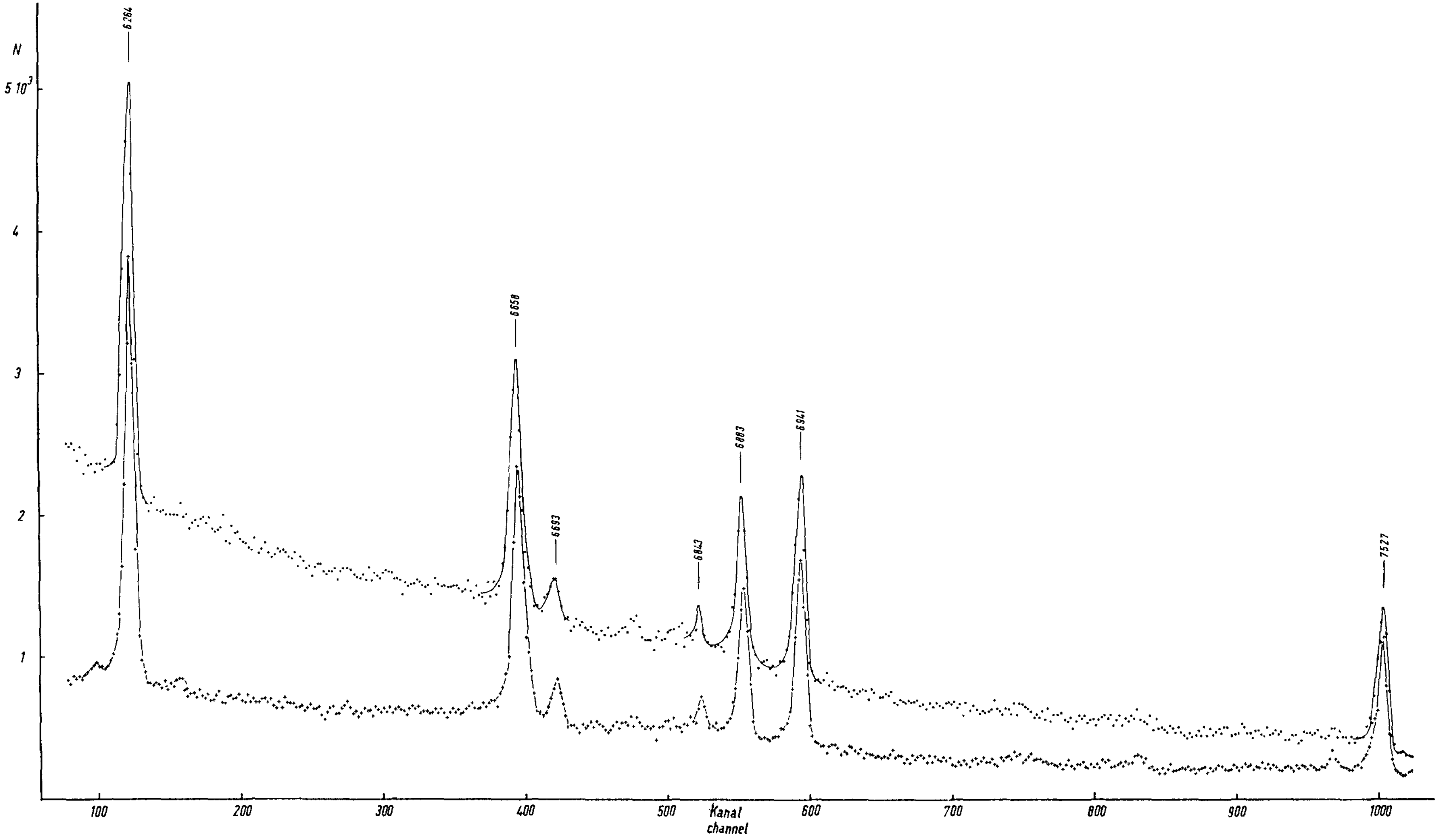

Fig. 4. Singles gamma-ray spectrum from thermal neutron capture in natural strontium carbonate observed with a 2.8 cm^2 x 0.2 cm Ge(Li) detector. Energy range from 6264 keV to 7527 keV. Reduction of background by application of pulse shape discriminator (lower curve) is clearly demonstrated.

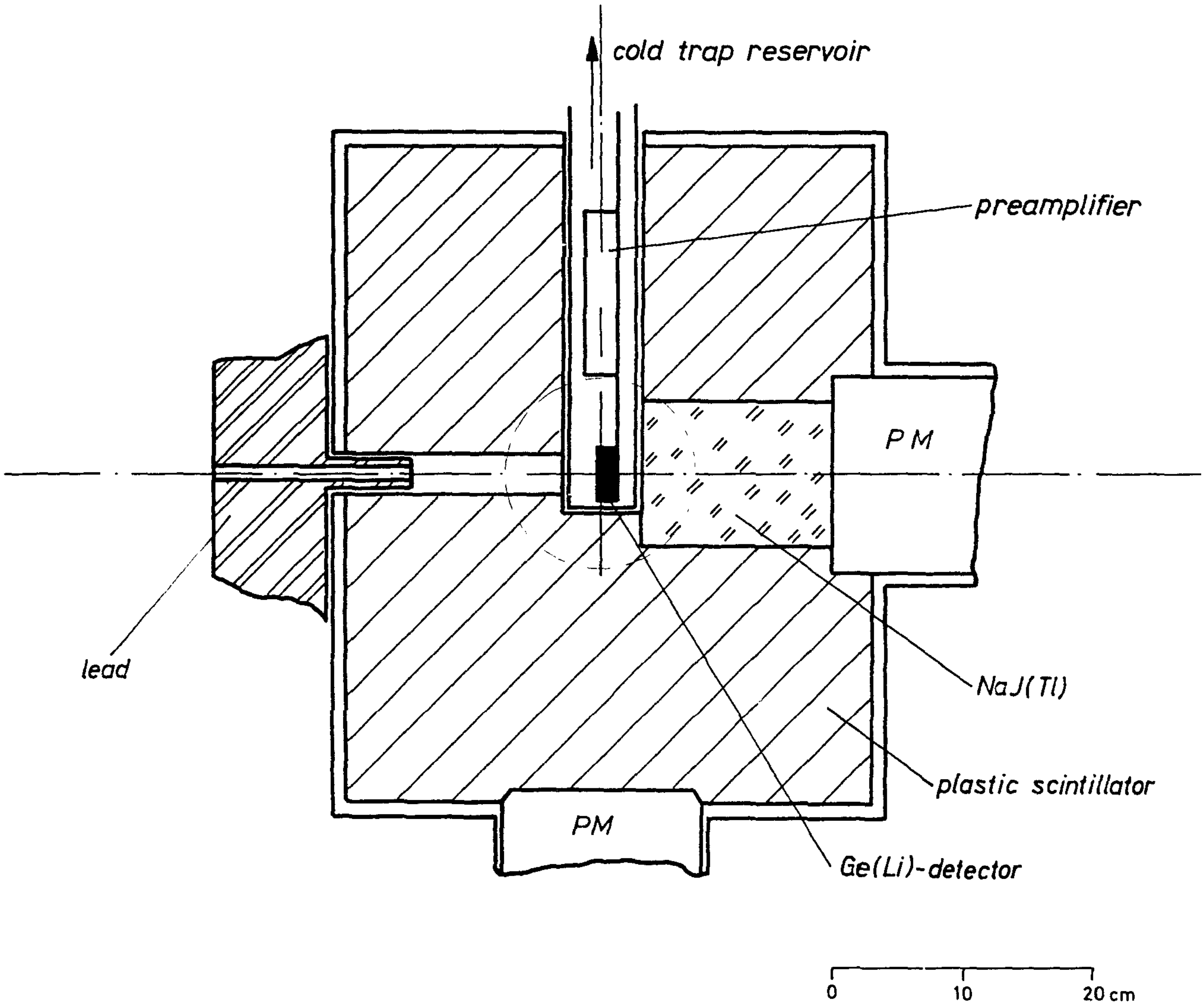

Fig. 5. Schematic of the general arrangement of anti-Compton spectrometer.

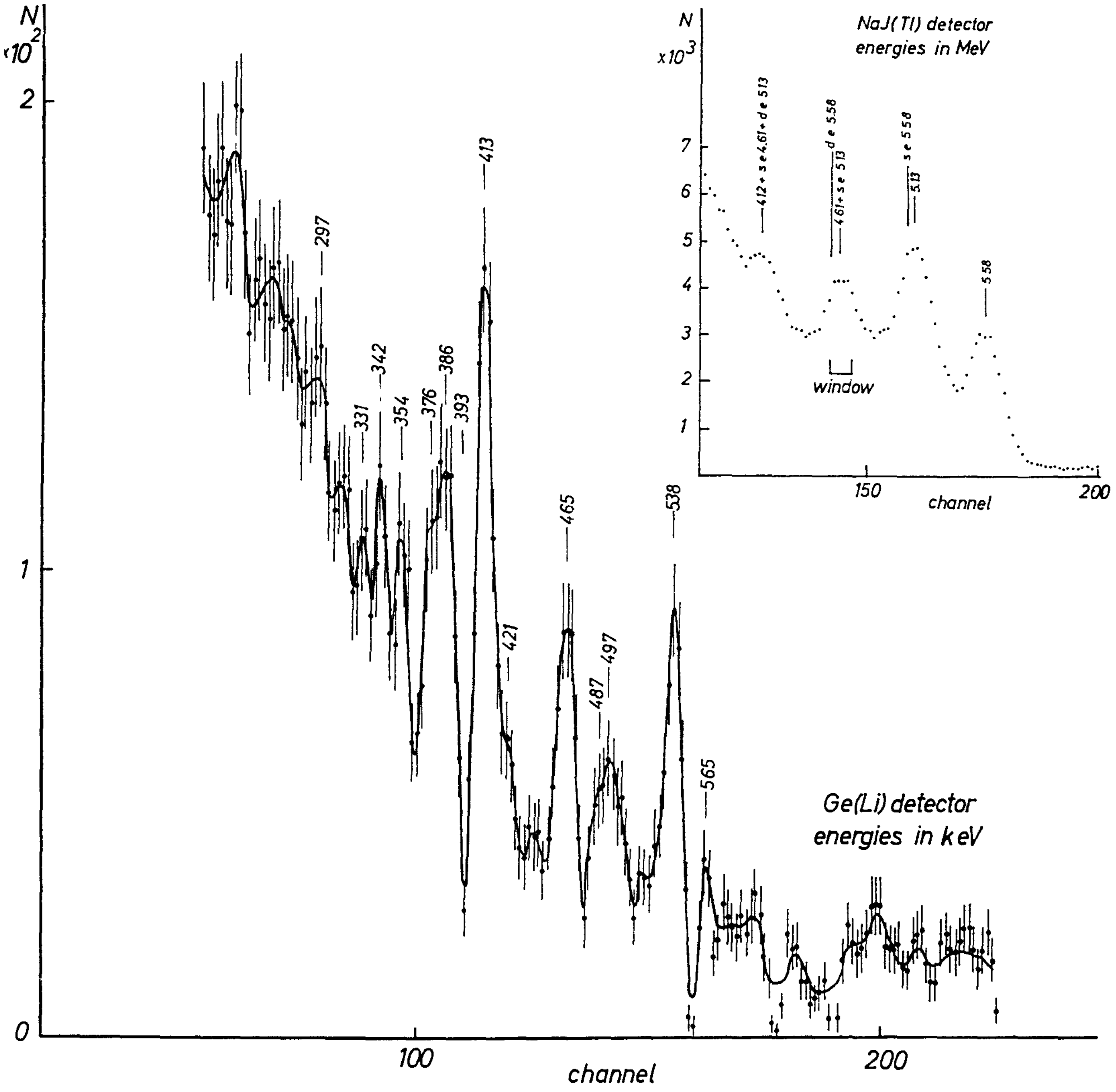

Fig. 6. Gamma-gamma coincidence spectrum of thermal neutron capture in Dy^{164} registered with a 4.0 cm^2 x 0.5 cm Ge(Li) detector and a 4"dia. x 5" NaI(Tl) crystal as gating counter. Window setting at 4.61 MeV as shown in the insert. The target was $Dy_2^{164}O_3$ enriched to 92.7 % Dy^{164}.