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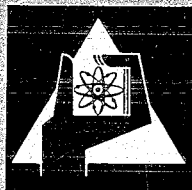
Angular Correlation Measurements on Gamma-Ray Cascades
following Thermal Neutron Capture

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Reprint from

"NEUTRON CAPTURE
GAMMA-RAY SPECTROSCOPY"

ANGULAR CORRELATION MEASUREMENTS ON GAMMA-RAY CASCADES FOLLOWING THERMAL NEUTRON CAPTURE

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Abstract

ANGULAR CORRELATION MEASUREMENTS ON GAMMA-RAY CASCADES FOLLOWING THERMAL NEUTRON CAPTURE. An experimental arrangement for angular correlation measurements on gamma-ray cascades following thermal neutron capture which is operated on-line to a computer is described. All of the interesting information can be accumulated simultaneously so that the determination of a large number of angular correlations is possible. Systematic errors arising from the coincident background under the peaks and from the complex structure of the capture spectrum can be eliminated either by a two-dimensional spectrum stripping procedure or, with sufficient accuracy, by application of the double-window technique. Practicable handling of data and comprehensive controls are thus possible. The extensive set of data obtained allows the results and their mutual consistency to be checked.

1. Introduction

Angular correlation measurements are of particular interest in experimental nuclear physics, because they supply information on characteristic properties of nuclear states that are necessary for the interpretation of nuclear level schemes and often constitute the basis for comparison with suggested nuclear models. Difficulties encountered in angular correlation measurements on γ-ray cascades following thermal neutron capture may arise mainly from the large number of γ rays usually present in the capture spectra and the high background coming from Compton scattered high-energy γ rays. In many cases, the capture cross sections are small and only small amounts of target material are available, thus requiring long measuring periods. Since the stability of the experimental equipment is limited, overly long measuring periods may also introduce errors, and one is forced to accumulate as much useful information as possible in a preset time. Another problem is the elimination of the coincident background underlying the peaks. This background is the main source of systematic errors in these measurements.

2. General Aspects

A difficulty common to all angular correlation measurements is the fact that the results can not always be explained unambiguously. In general, five unknowns are involved in a two-step cascade, namely the spins of the initial, intermediate and final levels and the multipole mixtures, δ, of the two γ-transitions. However, only the two coefficients A_2 and A_4 of the Legendre polynomials P_2, P_4 in the correlation function

$$W(\Theta) = 1 + A_2 P_2(\cos \Theta) + A_4 P_4(\cos \Theta) \qquad (1)$$

can be determined. In eq. (1), $W(\Theta)$ is normalized so that $A_0 = 1$. Octupole radiation is assumed to be negligible.

In many cases, the number of unknowns can be reduced either by using additional information from other experiments or by suitable choice of the experimental conditions. In neutron capture spectra primary transitions play a special role. If s-wave neutrons are captured in a target nucleus with spin I_t, the two spin states $I_p = I_t \pm 1/2$ are formed in the product nucleus. For even target nuclei one gets the unique capturing state spin $I_p = 1/2$ and the spins of lower levels may be determined from the angular correlation of two-step cascades involving primary transitions. If the spin of the ground state is known, it is useful to measure the correlation of cascades beginning in the capturing state and ending in the ground state and to apply the well known sum coincidence technique. For such measurements, however, only cascades with intense primary transitions can be used. These transitions are of pure dipole, mostly E1, character $\diagup 1 \diagup$. Hence, $A_4 = 0$ and only one measurable parameter remains. An unambiguous spin determination is possible if the final state has a parity different from that of the intermediate state and if it can be assumed from the radiation intensities that the second transition also has predominantly dipole character. With odd spin target nuclei this method can be applied if the thermal neutron capture is determined mainly by a single resonance or by resonances with the same spins. But, in general, both possible spin states participate in thermal neutron capture. The extent of their contribution to a certain primary transition cannot be stated in advance, since the resonance parameters and the partial radiation widths required for the calculation are not known accurately enough. In the theoretical expression of the angular correlation another unknown parameter then occurs which describes the branching of the intensity of the primary transition between the two spin states. This precludes an unambiguous interpretation in many cases. Therefore, in even product nuclei it is often more advantageous to investigate the angular correlation of γ rays in cascade with the intense ground state transitions from the first or second excited 2^+ levels. These transitions have pure quadrupole character and the remaining two unknown quantities can be determined, in principle, by the measurement of one correlation. The price for this advantage is the greater complexity of the spectra taken in coincidence with low-energy γ rays and the higher background compared to the simpler spectra coincident with high-energy primary transitions.

The ambiguity of the information can also be removed by the measurement of a larger number of correlations, which partially involve the same levels and transitions. If, for example, in a triple cascade (Fig. 1) the spin of the lowest level is known, the six unknown quantities I_1, I_2, I_3, δ_1, δ_2, δ_3 can be determined from the three angular distributions:

$$W_{\gamma 1-\gamma 2}(\Theta) = W(I_1, I_2, I_3, \delta_1, \delta_2, \Theta)$$

$$W_{\gamma 2-\gamma 3}(\Theta) = W(I_2, I_3, I_4, \delta_2, \delta_3, \Theta)$$

$$W_{\gamma 1(\gamma 2)\gamma 3}(\Theta) = W(I_1, I_2, I_3, I_4, \delta_1, \delta_2, \delta_3, \Theta)$$
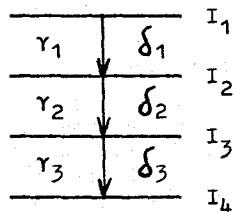

FIG. 1. Unknown quantities involved in a triple cascade.

The method applied to this simple example can easily be extended
showing that as many cascades as possible should be investigated.
Knowledge of their angular distribution also allows for valuable
checks and permits accurate corrections for perturbing coincidences.
Furthermore, a redundancy of the results is highly desirable in view
of the many possible sources of error. Since the measurements re-
quire a long time to accumulate meaningful statistics, the maximum
number of correlations of interest should be measured in one run.
The best way to fulfil this requirement and overcome the diffi-
culties is to perform a three parameter experiment with two para-
meters available for the energies of the γ rays in cascade and one
parameter for the angle defined by the detector positions.

3. Experimental Details

There were some other factors that determined the arrange-
ment of our equipment in the present form. One of these was the
intention to avoid systematic errors as far as possible and another
one was the need for the investigation of nuclei with small neutron
capture cross sections.

The setup of the equipment has been described in detail in
earlier work /̲2̲7. Only the main points will be repeated below. Fig. 2
is a schematic drawing of the arrangement installed at a horizontal
core channel of the Karlsruhe research reactor FR-2. Thermal neutrons
are diffracted from a lead single crystal in order to remove fast
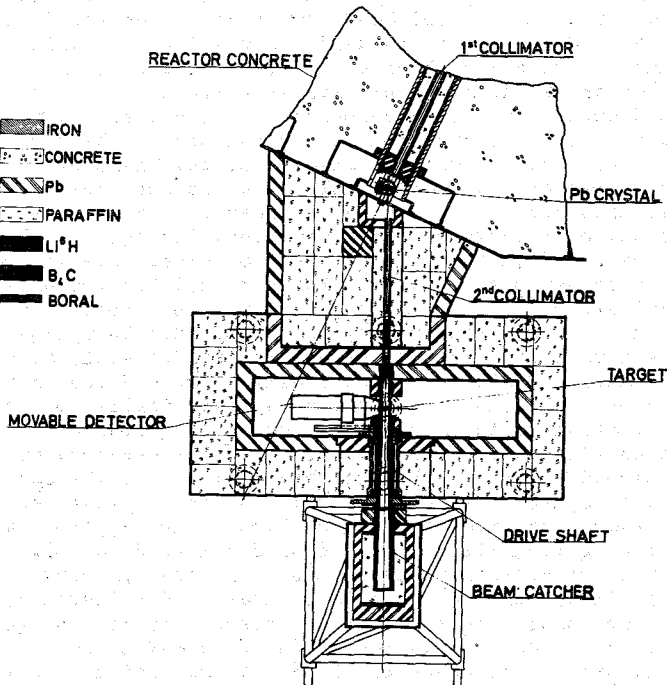neutrons and γ-ray contamination present in the initial beam. Up to now

FIG.2. Schematic drawing of experimental arrangement at the Karlsruhe research reactor FR-2.

the target was viewed by two gain-stabilized /3/ scintillation spectro-
meters with 4" dia. x 5" long NaI(Tl) crystals mounted in a plane per-
pendicular to the neutron beam, one detector being fixed in the verti-
cal direction. This detector can easily be replaced by a Ge(Li) diode.
The other detector can be rotated around the axis of the beam for a
range of angles from 60° to 360° with respect to the stationary de-
tector. To avoid systematic errors due to incorrect adjustment of the
source or of the beam and for purposes of control during the subse-
quent evaluation of data the movable detector is cycled through the
angular range of two quadrants. The electronics include conventional
coincidence circuits and sum coincidence circuits as well. The usual
fast-slow technique is applied with an effective time resolution of $2\tau =$
25 nsec in the fast coincidence stage.

        The entire system operates automatically. It is controlled
by a neutron monitor located in front of the target in the deflected
beam. A preselected number of neutron pulses determines the measuring
time in one angular position. At the end of the interval the control
counters are punched out, and the next angular position is set
according to the selected program. The typical measuring time in one
angular position is 30 minutes. Dead times of the equipment, expecially
those of the ADC, are taken into account automatically.

4. Data Acquisition

        Recording of data is done with the dual ADC coupled to the
Karlsruhe data acquisition system MIDAS (Multiple Input Data Acqui-
sition System) /4, 5/.

        As shown in Fig. 3, the heart of this system consists of
the coupled CDC 160 A and CDC 8090 computers, processor 1 and 2,
respectively. A disk storage is used for on-line totalizing up to
256 K channels with a length of 21 bits. A total of 68 K channels is
available for the angular correlation experiment and three subroutines
are at the disposal of the experimenter. In the first subroutine the
digital pulse height information is accumulated on the disk in a
256 x 256 channel matrix without any reduction. Whenever the angle
between the detectors is changed, the data are read out automatically
on magnetic tape and the disk area is cleared. Further data processing
is carried out on a large off-line computer. Storage in full reso-
lution allows for application of a two-dimensional spectrum stripping
procedure which has been proved to be most suitable for off-line back-
ground elimination /6/. In most cases, a more direct processing is
preferred which can be achieved by use of the double window technique
/7/. This method is not as rigorous, but sufficiently accurate re-
sults are obtained faster and more easily. Fig. 4 demonstrates the
application of the method. In order to systematically correct for the
spectrum coincident with background underlying a peak of interest two
or even three windows are needed. One window is centered on the peak,
while the second and third windows, having in total the same width as
the first one, select pulses of slightly higher or lower energy which
might come from Compton scattering of higher-energy gamma rays or
form interfering transitions (cf. below for more details). As shown
in the figure the contributions of the individual primary γ rays to
the coincidence spectra are clearly separated. For instance, in the
coincidence spectrum with the 6.96 MeV transition none of the lines
which are in coincidence with the strong 7.26 MeV transition appears
while in the singles spectrum the 6.96 MeV transition is only a very
small bump. The technique requires a second MIDAS subroutine which
allows digital windows to be set on the energy axes. The coincident
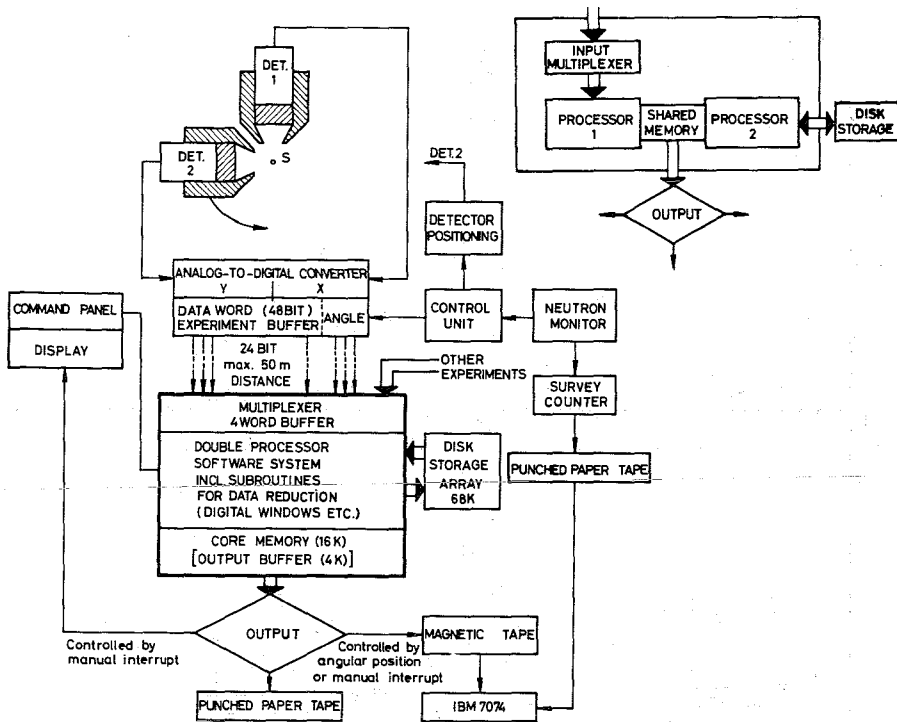pulses are routed into subsections of the disk according to these

FIG. 3. Simplified block diagram of the data-acquisition system.

window settings and according to the angles defined by the detector
positions. At present, a maximum of 32 windows can be set in each
energy direction and 1024 channels are available for each spectrum.

Each section or group of sections of the disk can be dis-
played on the CRT, dumped on magnetic tape or punched out by manual
interrupt so that a full evaluation of the state of the experiment
is possible at any time. The presence of significant anisotropy is
identified immediately (Fig. 5).

5. Off-line Data Processing

Off-line computer processing of the raw data consists of the
following operations (Fig. 6):

1) Sorting out of faulty data.

2) Gain shift control (Corrections for small shifts in gain
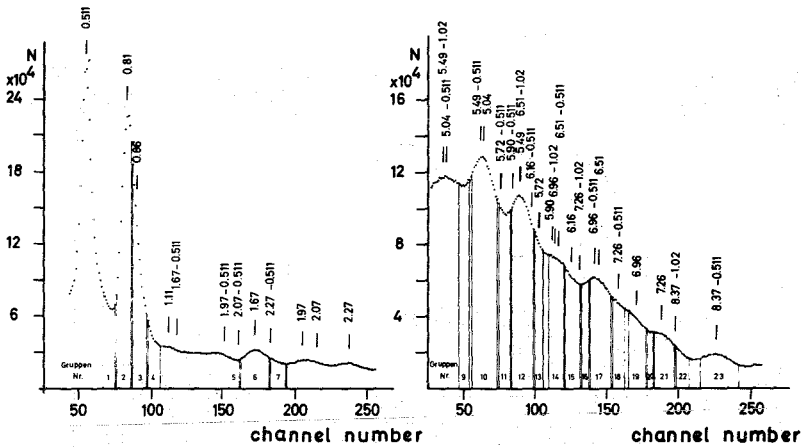and base line are made if necessary).
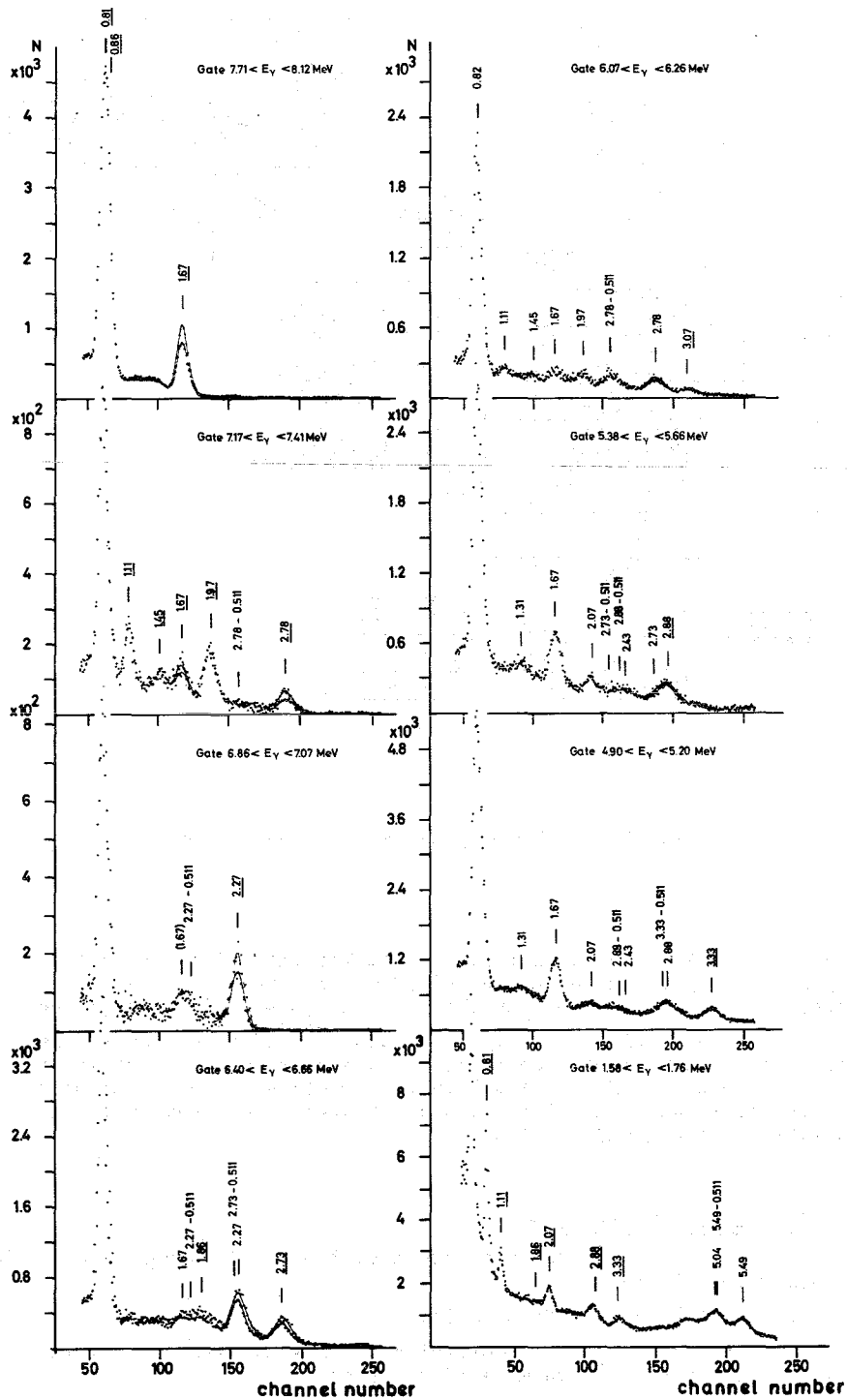
FIG. 4. ^{57}Fe(n, γ)^{58}Fe coincidence spectra at 90° and 180°. The coincident background is subtracted in each spectrum. Window settings in the spectrum of the fixed detector are shown above.
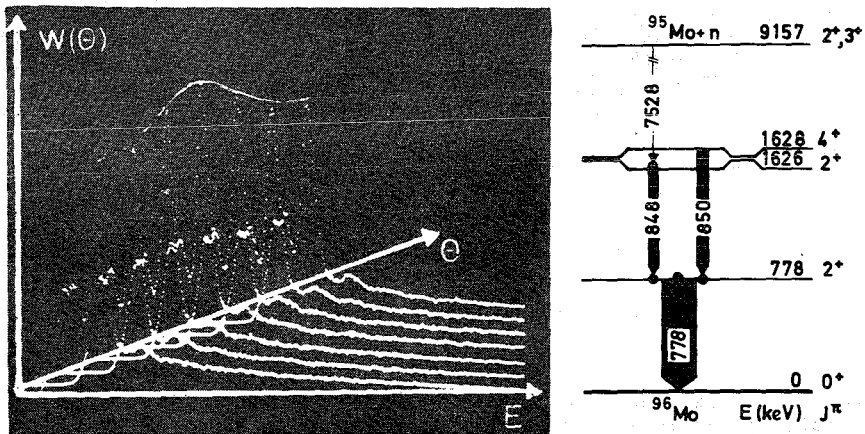

FIG. 5. Angular correlation of the (848 + 850) - 778 keV cascade from ^{95}Mo(n, γ)^{96}Mo displayed on CRT. NaI(T1)-detector spectra taken at 90°, 120°, 150°, 180°, 210°, 240° and 270°. Digital window settings in the spectrum of the second detector at 850 keV. The relevant part of the ^{96}Mo decay scheme is shown.

3) Channel by channel summing up of all coincidence spectra pertaining to the same angles (not necessary if the data are totalized on the disk).

4) Normalization of the spectra to the individual counting rates of detectors 1 and 2.

5) Subtraction of accidental coincidences measured separately.

6) Two-dimensional spectrum stripping and determination of intensities, or correction for coincident background by application of double window technique.

FLOW CHART OF DATA ANALYSIS

FROM ON-LINE COMPUTER

PARAMETERS OF LINE SHAPE / CHANCE COINCIDENCES / SINGLES COUNT RATES / RAW DATA / COEFFICIENTS OF INTERACTION PROCESSES OF Y RAYS AND NEUTRONS / DIMENSIONS OF DETECTORS AND SOURCE

SORTING OUT OF FAULTY DATA — not o.k. → REMOVE DATA

o.k.

GAIN SHIFT CONTROL

SPECTRA TOTALIZED ON THE DISK

SPECTRA TAKEN SEQUENTIALLY AT THE DIFFERENT ANGLES

CHANNEL BY CHANNEL SUMMING UP

PLOTS

NORMALIZING TO SINGLES COUNT RATES

PARAMETERS OF BACKGROUND AND LINE SHAPE

SUBTRACTION OF CHANCE COINCIDENCES

DETERMINATION OF SUBTRACTION FACTORS

TWO DIMENSIONAL SPECTRUM STRIPPING AND DETERMINATION OF INTENSITIES

SUBTRACTION OF COINCIDENT BACKGROUND BY USE OF DOUBLE WINDOW METHOD

RESULTING SPECTRA   PLOTS   RESULTING SPECTRA   PLOTS

DETERMINATION OF INTENSITIES

FITTED SPECTRA   PLOTS

LEAST SQUARES FIT OF W (θ) TO DETERMINED INTENSITIES

PLOTS

MODIFICATION OF ANGULAR CORRELATION COEFFICIENTS

$A_2, A_4, \Delta A_2, \Delta A_4$
$x^2, D = N_{kk'}^{-1} (\sigma_k, \sigma_{k'})$

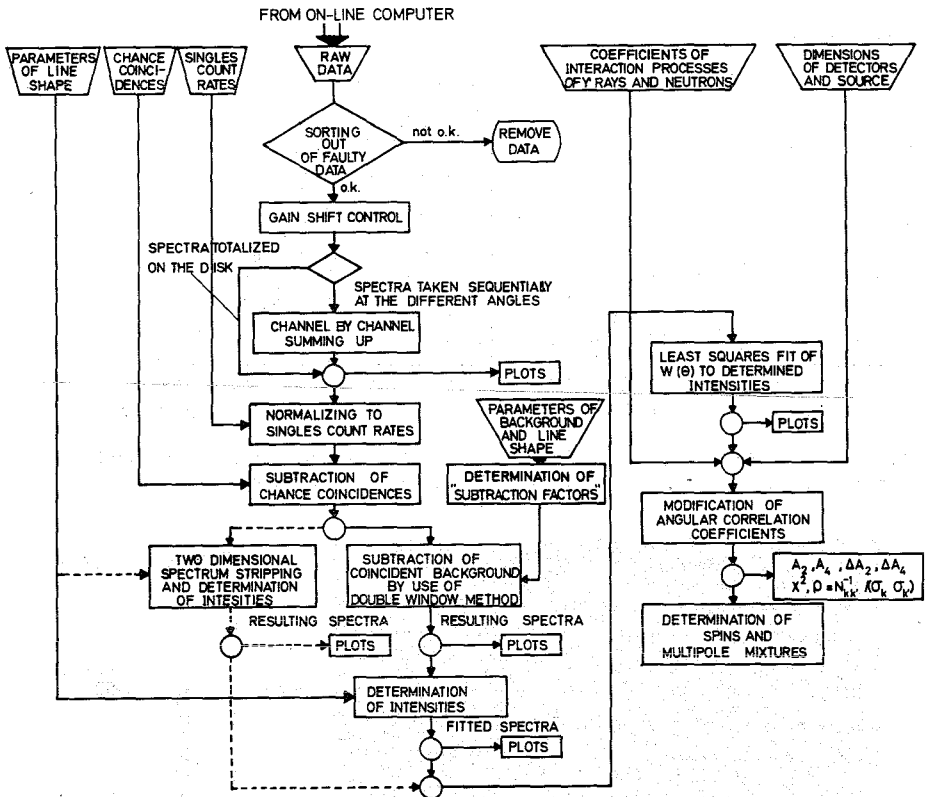DETERMINATION OF SPINS AND MULTIPOLE MIXTURES

FIG. 6.   Schematic diagram showing the off-line data handling.

The two-dimensional spectrum stripping procedure requires large computer programs and relatively long computing times. It has not been used for the evaluation of meaningful measurements up to now.

In order to correct for the coincident background by using the double window method the background coincident spectra are multiplied by appropriate factors and then subtracted from the spectra coincident with the selected peaks. The main problem of the method is the determination of these factors. They come to a first approximation from the window widths. Their exact values must be determined in a further computing step taking into account the individual shape of the spectrum and the nonlinearity of background. A check for the correctness of this procedure is the disappearance of a line not in coincidence with the selected transition. In order to get an estimate of the errors introduced by uncertainties in the background spectrum – particularly if there is no other check possible – the subsequent analysis is done also with spectra resulting from the application of different "subtraction factors" which follow from an upper and lower limit of the intensity of the background. These limits are determined

from the spectrum shape. For further processing of the resulting spectra
a computer program $\sqrt{8}\sqrt{}$ is employed which allows the analysis of
complex structures. An important factor in the analysis is the knowl-
edge of the γ-ray energies and their relative intensities which is
obtained from the singles spectra recorded with Ge(Li)-detectors. It
is only in this way that complex peaks in the NaI-spectra can be de-
composed unambiguously and correct estimates of the contributions
of single lines to a measured angular distribution obtained. A meas-
ure of the intensities of the γ transitions is the area below the
photopeaks. The calculation of this area with the computer program is
based on the assumptions that the line shapes are Gaussian and the
energy dependence of the background is a negative exponential function.
The parameters of the approximation function are automatically deter-
mined by the program. Fig. 7 shows, as an example, the analysis of a
doublet in ^{68}Zn. The intensity errors are compounded from the statis-
tical errors of counting, contributions coming from errors in the
goodness of fit and the uncertainties in determination of background
underlying the measured peaks. In favourable cases the attainable
accuracies are between 0.5 % and 1.0 %. As shown in the flow diagram
(Fig. 6) data analysis is supervised by means of plots which are
generated after each critical computer step. These plots are of great
value, particularly for the intensity analysis since it may happen
that the least squares fitting procedure of unfolding the spectra
might fail to obtain the absolute minimum of χ^2. In this case an
iterative procedure is used to repeat the analysis with varied in-
put parameters for line shapes and background. As usual the inten-
sities are fitted with the angular correlation function $W(\theta)$ $\sqrt{}$eq.(1)$\sqrt{}$
using the method of least squares $\sqrt{9}\sqrt{}$. Since the coincidence
spectra are taken in at least four angular positions, it is possible
to check the goodness of fit by a χ^2-test. If there are indications
of errors beyond the ordinary statistical uncertainties, the errors
associated with the correlation coefficients A_k are increased accord-
ing to the description given by Rose $\sqrt{9}\sqrt{}$. The uncertainty in the
determination of the background is also taken into account. The
correlation coefficents are corrected for the effects of the finite
size of the source and the detectors, and the interaction processes
of the gamma quanta and of the neutrons in the source, using the
methods of Michaelis $\sqrt{10}\sqrt{}$ and White $\sqrt{11}\sqrt{}$. If there are contri-
butions of interfering lines, e.g. single or double escape peaks of
higher-energy γ rays which can neither be resolved by the spectrometers
nor be taken into account by application of the computer programs the
correlation coefficients obtained have to be corrected for these contri-
butions in a final step.

          Spins and multipole mixtures are determined from the angular
correlation coefficients by application of well-known analytical and
graphical methods. For the analysis of correlations with one mixed
transition the method recommended by Coleman $\sqrt{12}\sqrt{}$ has proved to be
most convenient. This method allows the representation of a larger
number of results in one diagram thereby giving a good idea of the
precision of the measurements. The method is also used for the anal-
ysis of correlations with two mixed transitions. With the aid of a
computer and an incremental plotter it is easy to plot the necessary
series of ellipses each of which represents the variation of one mix-
ture while the other is fixed at a particular value. In other cases,
a new procedure $\sqrt{2}\sqrt{}$ is applied which is developed from the method
described by Poletti and Warburton $\sqrt{13}\sqrt{}$.
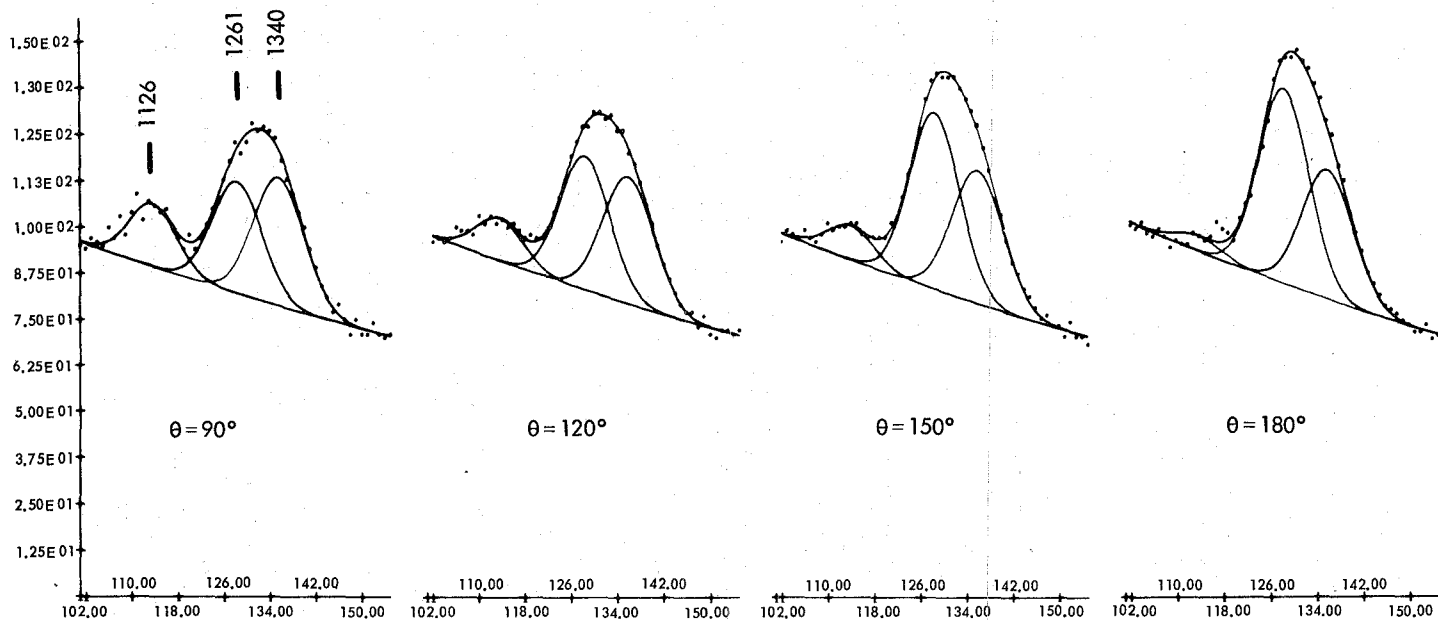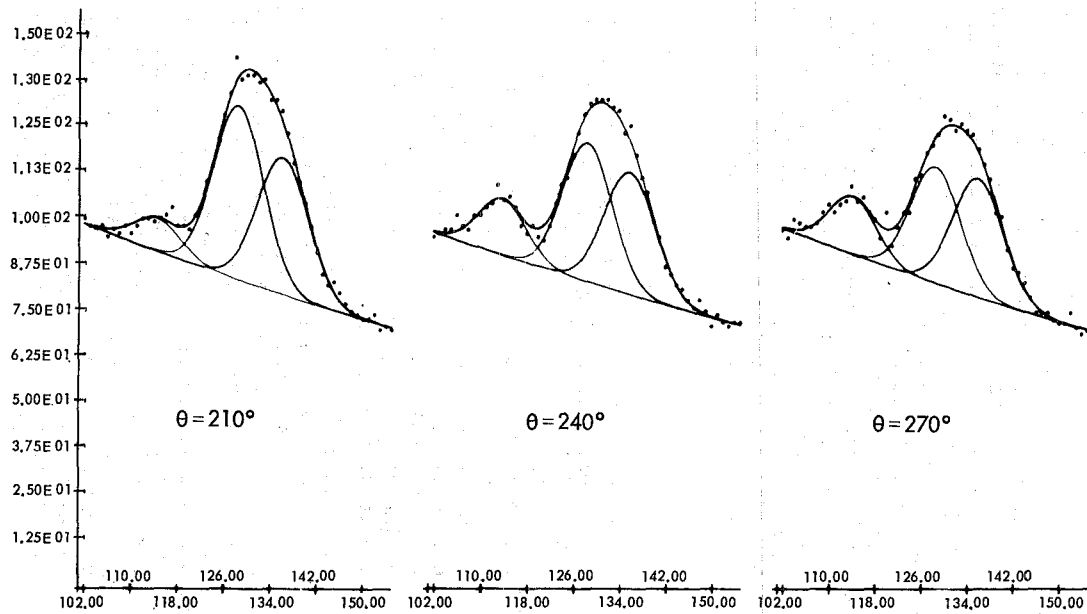
FIG. 7. ^{67}Zn(n, γ) ^{68}Zn angular correlation analysis of the complex peak at 1300 keV [18].
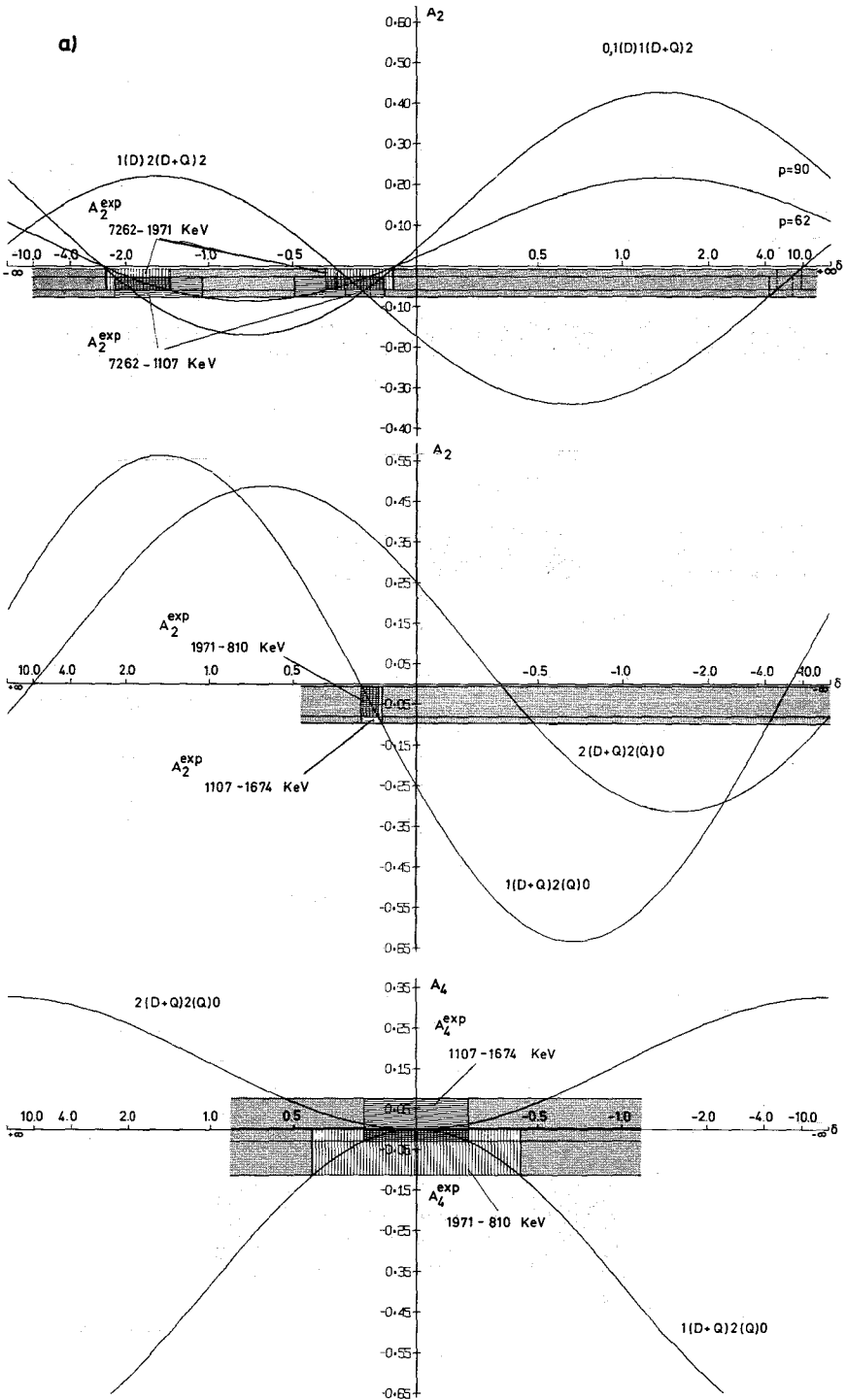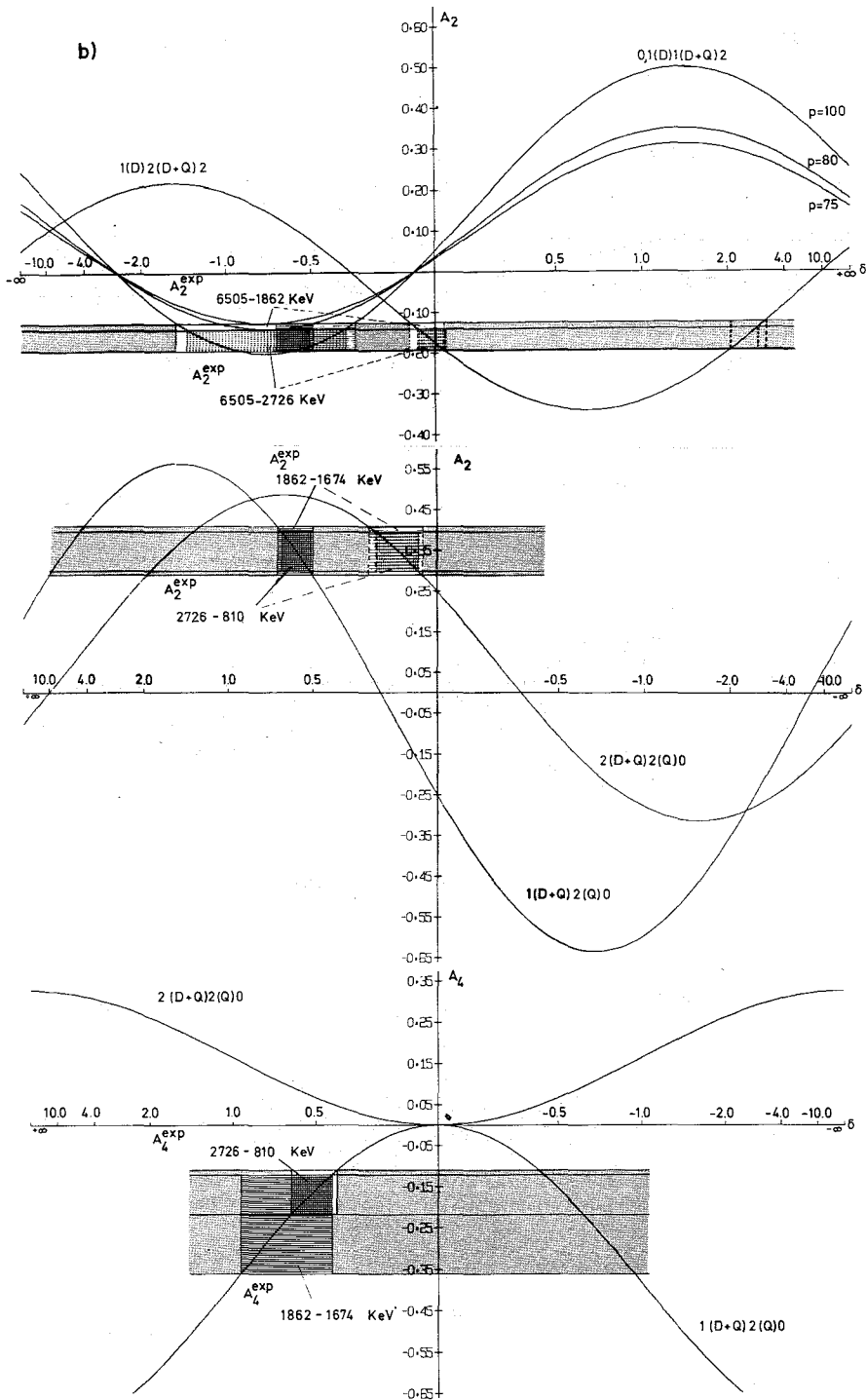
6. Test Measurements

The equipment was carefully tested for mechanical asymme-
tries, e.g. eccentricity of the target position and eccentricity and
inhomogeneity of the neutron beam. With various γ-ray sources the
change in counting rate at the different angles was in general less
than $\pm$ 0.5 %. As a further test, the angular distribution of the
2.38 - 0.84 MeV cascade from the reaction $^{32}S(n,\gamma)^{33}S$ was investigated.
This cascade goes through an intermediate state with spin 1/2 and hence
has an isotropic distribution. The correlation measured with a sample
of 7.13 g of pure sulfur in a sample holder of 1.8 cm diameter and
1.9 cm length resulted in values of the corrected angular correlation
coefficient of A_2 = 0.002 $\pm$ 0.004 and A_4 = 0.008 $\pm$ 0.012. Thus, the
distortion of the angular distribution by mechanical asymmetries can
generally be regarded as small relative to the other sources of error.
The equipment was tested and the program checked in a further series
of measurements using ^{60}Co sources of various diameters. The corre-
lations observed gave the expected results for the correlation func-
tion which is known to be $W(\Theta) = 1 + 0.102\ P_2(\cos\Theta) + 0.009\ P_4(\cos\Theta)$.
With a source of 2.6 cm diameter and 2.7 cm length the correlation
function obtained was $W(\Theta) = 1 + (0.107 \pm 0.006)\ P_2(\cos\Theta) + (0.009 \pm 0.015)\ P_4(\cos\Theta)$ showing that proper corrections were applied by the
program.

7. Results

Some of the results obtained from angular correlation
measurements on γ-ray cascades following thermal neutron capture in
^{57}Fe, ^{61}Ni, ^{67}Zn, ^{95}Mo and ^{97}Mo are presented in these proceedings
$/17$ - $20/$. From the systematics of even mass nuclei between A = 50
and A = 100 it is expected that the low energy levels of these nuclei
are adequately described by the vibrational model. Recently reported
shell-model calculations $/14$ - $16/$ have also been very successful
in predicting correct level energies. In order to clarify the level
structure, knowledge of the multipole mixtures of the transitions is
of great importance. Therefore, angular correlation measurements are
particularly valuable in this mass region.

Our investigations $/20/$ on γ-ray cascades following thermal
neutron capture in ^{57}Fe have been selected as an example to demon-
strate the efficiency of the method. In this case, more than 40 corre-
lations have been measured, 33 of which have been evaluated. An ex-
tensive set of results was obtained which allowed the mutually con-
sistent determination of a large number of level spins and multipole
mixtures. Some of the results are shown in Fig. 8 (Part of the decay
scheme relevant to the angular correlation measurements under dis-
cussion is shown in Fig. 9). The upper row in Fig. 8 represents the
analysis of cascades involving a primary transition. The capturing
state of ^{58}Fe is an unknown mixture of 0^- and 1^- states. P is the
parameter indicating the relative contribution (in %) of the spin 0
states to a particular primary γ ray. The mixture parameter δ of the
second member of these cascades is taken as the abscissa; the angular
correlation coefficients are the ordinates. In all cases, the spin of
the final state is 2. It can be concluded from the intensities that
the primary transitions have an E1 character. Therefore, the following
three alternative spin sequences are possible 0 - 1 - 2, 1 - 1 - 2,
and 1 - 2 - 2. The sequence 1 - 0 - 2 was ruled out because of the
nonzero anisotropies observed in these measurements. The middle and
bottom rows of the diagrams refer to cascades with ground state transi-
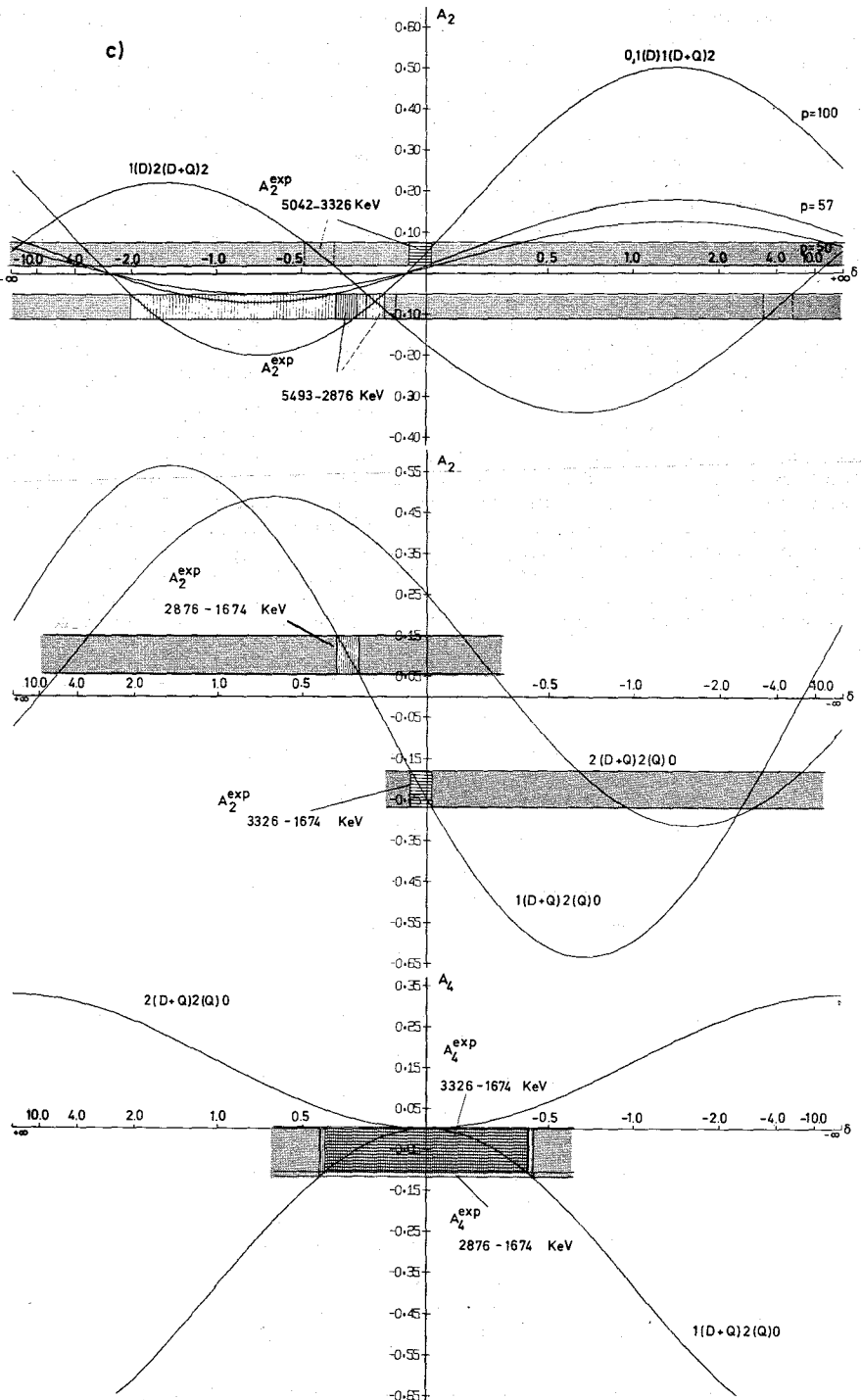tions. The first member of each of these cascades is the same γ ray

FIG. 8. ^{57}Fe$(n, \gamma)^{58}$Fe angular correlation measurements. Hatched bands represent the experimental results. Change in the sign of δ has been taken into account (cf. text).

as the second member of the cascades in which primary transitions are involved. With the definition adopted for $\delta\sqrt{21\_7}$ a change in sign of δ is necessary because of the different γ-ray order in these cascades. This has been taken into account by reversing the sign of the abscissa in the middle and the bottom rows. The observed γ-ray intensities and the neutron orbital momenta known from (d,p) reactions restrict the possible spin sequences in the low-energy cascades to 1 - 2 - 0 and 2 - 2 - 0. The spin 1 assignment of the 2782 keV level and the branching of the 7262 keV transition between the two capture state spins is determined from the angular correlation of the 7262-2782 keV two-step cascade to the ground state (Fig. 9). This branching (p = 76$\pm$14) and the spin assignment is adopted to determine the multipole mixtures of the 1971 keV and the 1107 keV transitions from the 7262-1971 keV and the 7262-1107 keV correlations, respectively. The results obtained are in excellent agreement with the values found from the analysis of the 1971-810 keV and 1107-1674 keV correlations (Fig. 8a).

It was not possible to determine the branching percentages of the 6505 keV, the 5493 keV and the 5042 keV transitions (Fig. 8b and 8 c) directly through a two-step cascade to the ground state. These branchings were determined from the angular correlation measurements taking into account the mixture parameter δ of the second member of each cascade and the spin assignments of the intermediate states which are known from the analysis of the low-energy cascades to the ground state. In this way, for about 70 % of the observed intensity, the contributions of the two capture state spins (0⁻, 1⁻) to the intensities of the primary transitions have been accounted for.
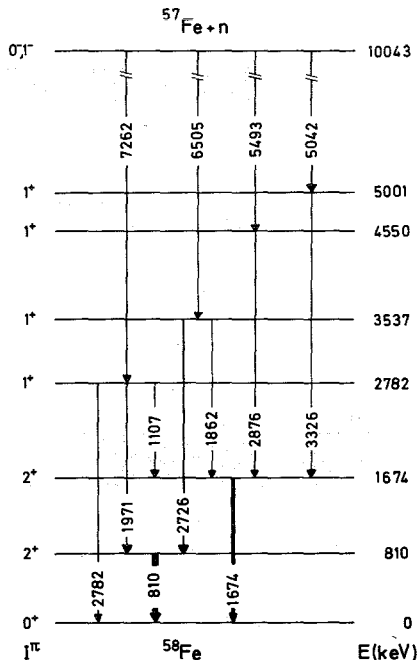

FIG. 9. Partial decay scheme relevant to the angular correlation measurements shown in Fig. 8.

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