[3] DOLNICAR, J., Consultants Meeting on Evaluation of Gamma Spectra and the Preparation of Programme for IAEA Training Courses on Gamma-Ray Spectroscopy, Vienna, 1988 (internal report).

[4] Determination of the Lower Limits of Detection and Decision for Ionizing Radiation Measurements - Part 1: Fundamentals and Applications to Counting Measurements without the Influence of Sample Treatment, Rep. ISO/DIS 11929-1 (TC 85), International Standards Organization, Geneva (1994).

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3. SOME DEFICIENCIES AND SOLUTIONS IN GAMMA RAY SPECTROMETRY[5]

3.1. SUMMARY

A number of problems in high-resolution gamma ray spectrometry as well as some deficiencies of existing computer programs for the quantitative evaluation of spectra are discussed and some practical solutions are proposed.

3.2. INTRODUCTION

In discussing a high-resolution gamma ray spectrum we usually make several implicit assumptions. For example:

(1) The signals coming from the linear analog electronics connected to the detector are converted into digital numbers where the value of the number is linearly proportional to the height of the analog signal.

(2) These numbers are used by a multichannel analyser to address elements in an array of memory words and to perform an "increment-by-one" sequence in the addressed word. The array of memory words is called a spectrum and the individual memory words are called the spectrum channels. There is an almost linear correlation between the channel number where an event is counted and the registered photon energy.

(3) In the graphical representation of a spectrum as shown in Figure 1, the abscissa is divided into bins of energy intervals (the channels) which are of equal width in terms of energy and which are defined in integer increments. All events whose energy falls into the interval from the start to the end of a bin will be defined as the count rate in that channel. The ordinate lists the number of counts per channel, i.e. the count rate accumulated during the measuring time. The display of a spectrum should be either a dot display, where the dots are positioned at the centres of the channels or, preferably, a histogram display where a horizontal bar marks over the total width of the bin the number of counts in the channel. The horizontal bars in adjacent channels are connected by vertical lines at the channel boundaries.

This section will address typical mistakes which may occur in the measuring of the spectra and which are sometimes overlooked as well as selected topics concerned with the analysis of spectra.

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[5] *Prepared by W. Westmeier, Gesellschaft f. Kernspektrometrie, Germany.*

3.3. ERRORS IN SPECTRUM MEASUREMENT

3.3.1. Pole-zero settings

The improper setting of the P/Z correction circuitry in the linear amplifier may lead to severe peak shape distortion, which may even show up as satellite peaks on either the low- or the high-energy side of the main peaks. In these cases, the peaks are analyzed as having shoulder or satellite companions which fill the excess number of counts in improper locations, and the residua found after peak analysis will indicate several additional peaks in the flank of the main peak. One can in principle invent and fit peak shapes which take into account such poor P/Z settings, i.e. peak shape functions which include low- or high-energy tailing contributions to the respective analytical shape. However, these procedures disable the program to detect and analyse real shoulder peaks, which might be found close to major components, as these then will also be taken as "P/Z satellites". It is therefore highly recommended that spectra be measured with properly set P/Z circuitry in the spectroscopy amplifier. Operator control of the P/Z setting with an oscilloscope has been proven to be the best suited most reliable and most economical procedure.

3.3.2. Measurements at high count rates

In measurements at very high count rates one will often find the problem that random pileup processes become dominant and the shape of the peaks is significantly distorted. There are amplifiers on the market, which can correct (disable) for pileup events, but in our experience, these electronic methods may lead to other effects which complicate the quantitative analysis of the spectrum such as an incorrectly defined lifetime correction. It is therefore recommended that count rates should be kept so low (<10 kcps) that there is no significant random pileup encountered. This can be achieved by a larger distance between the sample and the detector, by the choice of a shorter shaping time in the spectroscopy amplifier, or sometimes even through modifications in the experimental set-up. One can, for example, reduce significantly the number of random pileup events encountered in a measurement with an n-type semiconductor detector by placing a 0.3 mm thick cadmium cap over the detector. This absorbs essentially all low-energy gamma- and X rays and reduces the count rate significantly. In addition, it is a very effective means of avoiding the X ray/gamma ray true coincidences that make quantitative analyses with these detectors very difficult. If one cannot avoid measuring spectra at very high count rates, one should either use electronic correction circuitry which allows the consideration and correction of distorted events ("Loss-Free Counting") or, even much better, one may use extremely fast signal processing technology which is now available. The latter unit is called "Analog-to-Digital Signal Processor" (ADSP) and it works in principle as follows: A very fast 12-bit sampling ADC directly converts the preamplifier output signal every 50 ns. The digitized signal shape goes on-line into an array of signal processors which analyze, correct (e.g. for ballistic deficit) and permanently calculate the events. The digitally corrected result of the integration (= the channel number) is directly fed into the multichannel-analyzer (MCA). The advantages of the ADSP unit lie in the abandoning of disturbing and "slow" analog electronics, in the extremely high count rate throughput, in the intrinsic corrections for distorted signal shapes, in the very effective pileup suppression (400 ns pulse pair resolution), and the specification that there is essentially no ADC deadtime. The spectrum broadening is reported to be less than 10% change in FWHM for the 1332.5 keV line of ^{60}Co at 1.5 µs shaping time, 300 kcps input rate and 30% throughput. The spectrum shift under these conditions is less than 100 ppm.

3.3.3. Odd–even effects

It is surely a rare event but nevertheless still seen that the contents of counts per channel show a significant odd-even effect (Figure 1).

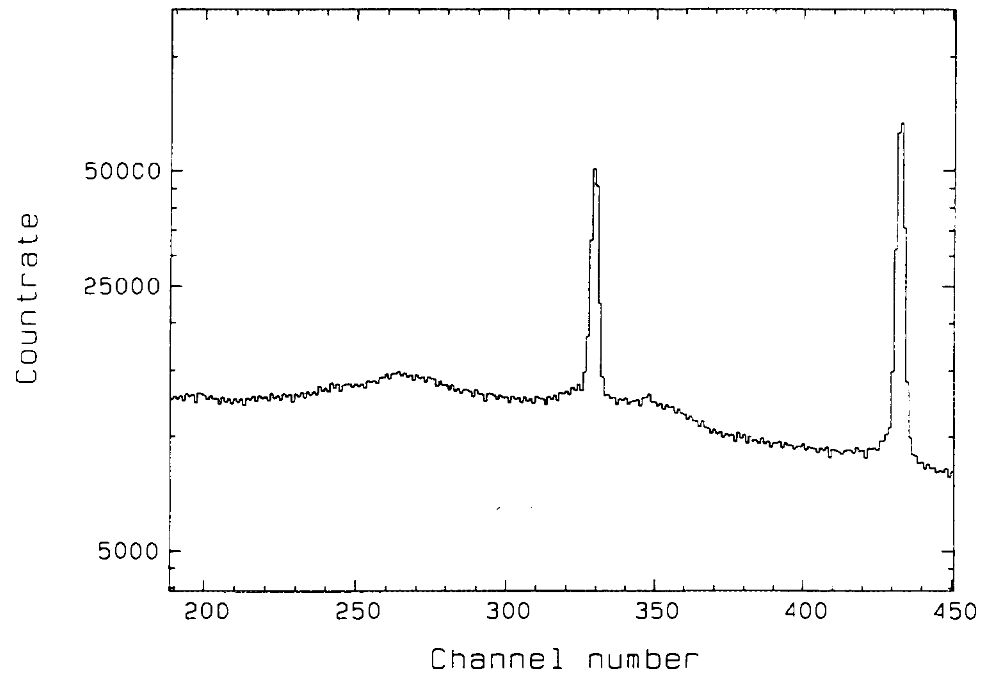

FIG. 1. Example of a spectrum with a pronounced odd-even effects in every second channel.

This feature, in which channels in binary increments show excessive or missing count rates as compared to the other channels, is caused either by a very poor differential linearity in the ADC or very rarely through binary transfer errors between the ADC and the MCA. In the more frequent case of odd-even effects through a poor differential linearity, all events measured by the detector do appear in the spectrum, however, some of them are in a systematically placed in the wrong channel number. Thus, the area of any peak analysed will be almost correct, but the residuals and correspondingly the uncertainties of the calculated results will be unacceptable. Normally these odd-even effects are easily detected as regular patterns upon visual inspection of peak-free areas of the spectrum.

3.4. COMPUTER-AIDED SPECTRUM EVALUATION

In the first years after the boom of personal computers in 1981, the number of computer codes for the analysis of gamma ray spectra grew tremendously and one could find essentially one gamma-spectrum code per laboratory. This situation was unsatisfactory and puzzling because one could not find a widely accepted program that might serve as reference for intercomparison purposes. The situation has been significantly corrected in the last years through the effort of international organizations or other authorities to find acceptable criteria which may serve for intercomparison purposes and which may qualify and rank the codes, so that at present, there are less than 40 codes on the market which are in an open competition for user's interest. Some of these codes, however, are highly specialized for certain applications and should not be counted as general purpose gamma ray codes. This situation when only few, well tested codes are available seems more favourable, but there is a big "caveat" for users: Modern gamma spectrum analysis codes are very complex programmes in which very many procedures and tasks are implemented through machine-oriented programming tools. These computational procedures allow the very fast execution of very complex calculations or decisions, but they also allow the very fast propagation of complete nonsense. Thus, one still has to find methods and procedures that help to control these highly complicated programs. One has to make sure that the listed results are in fact in agreement with the results to be expected and that the uncertainties assigned to the calculated results are in conformity with the accepted rules and definitions.

There are several problem areas in which computational, algorithmic or logical errors may occur and where the results output by programs may need to be looked at very carefully. Some of the more common of these are described below.

3.4.1. Gamma ray/X ray mix

Whenever a gamma ray spectrum covers the energy range below ca. 80 keV one will also find the X ray peaks from activities assayed as well as fluorescence induced X rays from the detector construction and surrounding material. In most cases one simply ignores the X ray peaks in the spectrum because these cannot be correctly analysed with a gamma ray analysis program. The peak shape of X rays is by principle different from the shape of gamma ray peaks; it is a Voigt function, i.e. a function with a significantly widened peak base, and this peak-shape is normally not considered by gamma ray analysis codes. As a consequence one will never find correct results when one analyses overlapping peaks in the X ray regime above ca. 30 keV with a gamma ray code. One can, however, easily analyse a gamma ray singlet peak, which has no considerable overlap with other X ray peaks. At energies below ca. 30 keV the Lorentzian contribution to the Voigt function is essentially negligible and one will get acceptable results when a multiplet of gamma ray and X ray peaks is simultaneously analysed. In these cases one has only to take into account that, due to the very close positioning and overlay of several X ray peaks from nearby subshell transitions, the shape of X ray peaks may be significantly different from the gamma ray peaks, i.e. the program must allow for different shape parameters of the individual overlapping peaks in one region.

3.4.2. Backgrounds in gamma ray spectra

There is often quite a bit of confusion when the term "background" is used in gamma ray spectrometry because there are different sources of counts which do not belong to the net full-energy peak area, which are denoted as background. Generally one should discriminate between the "external" and the "internal" background:

The external background is determined by those events, which are measured when no sample is placed in front of the detector. These include contributions from terrestrial and natural sources of radioactivity, cosmic radiation, induced X rays from construction materials etc. The external background is a spectrum with some continuum distribution and discrete full-energy peaks sitting on top. It should be very well determined for each measurement set-up. The external background spectrum, out of which the peak contributions are to be analysed, should be measured for a considerably longer time than any sample spectrum. All peaks in the external background spectrum must be quantified in terms of counts per unit time together with their corresponding uncertainties. In every sample spectrum one can then later subtract the contribution of the external background to the measured peak-areas with proper consideration of the statistically correct uncertainties. Any other method of pro-rata subtraction of an external background spectrum from a sample spectrum will lead to a resulting net spectrum with distorted statistics.

The internal background constitutes of those events other than full-energy peak counts, which are produced by the sample in front of the detector. Among these are the results of photon interactions with the detector material (Compton, ballistic deficit, and escape events, etc.) or with the surrounding material (backscattering, fluorescent excitation) and all related secondary events. Again, the internal background consists of a spectrum with some continuum distribution and discrete full-energy peaks. The internal background spectrum cannot be assayed and processed ahead and considered later in the analysis of the sample spectrum. Rather, one has to separate out the continuum part, analyse all peaks in the sample spectrum and define (quantify) the contributions from the internal background.

There are yet other events which are often denoted as "background" and which must be quantified and subtracted from the gross count-rate in order to find the net full-energy peak counts. These are the events underlying the full-energy peak, which come from Compton events of other peaks at higher energy, from full-energy counts suffering from ballistic deficit, or from other secondary interactions. In order to avoid confusion it is recommended that the distribution of these events under the full-energy peak should be denoted as ***baseline*** [6]. It has been noted long ago [3] that the shape of the baseline under full-energy peaks follows a step-function. In fact the baseline constitutes of two components, namely a smooth, almost linear Compton contribution from events at higher energies plus a step-function contribution which is caused by the events in the full-energy peak itself.

An algorithm has been published [15, 16] which allows to calculate the baseline distribution under singlet and multiplet peaks without prior knowledge of either the number of components in a multiplet or the shape of the peaks. The algorithm even enables one to calculate the correct shape of the baseline under escape peaks where the continuum distribution on the high-energy side of the peak sits at a considerably higher level than at the low-energy side (Figure 2). This "step-up" feature comes from the fact that three events are coincidentally involved in the pair production process. The primary photon energy minus 1022 keV (twice the electron mass) is found as kinetic energy of the electron and positron, respectively, and there are two 511-keV photons from the annihilation of the positron, which was originally created in the pair production process. Thus, because the kinetic energy of particles is always measured as a full-energy event (the double escape peak), there is an additional probability that all or part of the energy of another photon will be registered in coincidence and the summing event is measured in a higher channel number. The additional probability of such an event to occur gives rise to an elevated continuum distribution at the high-energy side of escape peaks. It should be noted that due to the above-described mechanism one will find the characteristic Compton distribution and one Compton edge for a 511-keV event above the double escape peak (Compton 1) and above the single escape peak (Compton 2), as well as the usual Compton distribution and Compton edge below the full-energy peak (Compton 3). In Figure 3 one can distinguish the characteristic features measured from high energy photons, where the Compton edge above the single escape peak is buried in this example under another small peak.

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[6] *The term "baseline" was proposed by K. Heydorn from Risø (Denmark).*

3.4.3. Length of spectra

With the availability of cheap computer memory and mass storage devices it has become fashion that spectra are measured with an ever-increasing number of channels. Whereas the first MCA units in the seventies had only 256 channels for a spectrum one can now find ADC and MCA units which allow to digitize and store spectra with up to 64k channels. The enhanced resolution in terms of keV per channel, however, does not automatically give better possibilities to resolve closely positioned peaks. In fact, it is very often a significant disadvantage when spectra are measured with an excessive number of channels. There is no generally applicable recipe, which allows one to define the optimized spectrum length, but there are several constraints that should be considered:

It is clear that the resolution of the detector is the most important variable, which defines the spectrum length. Most of the common non-linear least-squares algorithms used for the fitting of peaks are mathematically stable and yield reliable results when the full-widths-at-half-maximum (FWHMs) of peaks are in the range of 3 to 6 channels. This finding comes from the fact that most algorithms make point-fits, i.e. for the minimized function (Y_{exp}-Y_{calc}), the calculated function value Y_{calc} and the experimental number of counts in a channel Y_{exp} are both taken to be situated in the centre of a channel. This algorithm yields correct results as long as the FWHM of peaks is larger than ca. 3 channels because the shape of the peak function within the energy bin is then very well approximated by a straight line and the Centre-of-gravity of the function lies in the centre of the channel. If, however, the FWHM of peaks is significantly smaller than 3 channels, the curvature of the function within the energy bin must be considered and one may no longer apply a point-fit procedure. In a systematic survey we have shown that using an integrating fit which considers the curvature of the function within each channel, one can achieve a reliable and stable fit of peaks where the FWHM is as small as 0.9 channels. However, as most commercial spectrum analysis codes do not provide this integrating fit feature, it is recommended that the rule-of-thumb "the FWHM of peaks to analyse should be 3 channels or above" be applied. Nevertheless, one should try to keep the number of channels in the spectrum as small as possible!! Thus if one wants to measure peaks at 120 keV and above, and the detector resolution at 120 keV is 1.2 keV FWHM, then the spectrum should be measured with a resolution of 1200 eV / 3 channels = 400 eV/channel and the spectrum length is determined by the maximum energy assayed. As almost all essential gamma rays from nuclear decay are found at energies below 1500 keV it is normally sufficient to measure spectra with 4k channels (4096 channels) in length. The measurement of nuclear reaction gamma spectra with energies usually up to 10 MeV or above will require spectrum lengths of 8k or even 16k channels. The main reason for this recommendation of a spectrum as short as possible is the enhanced statistical significance of the data. When the same number of counts are distributed over 8k channels rather than 4k channels, then the statistical accuracy of the count rate in each channel is SQRT (8k/4k)=1.4 times worse and the fit algorithm will encounter less strict driving forces through the weights of the count rates in the channels. Therefore any analysis code will have problems in spectra which are measured over too many channels to find and quantify either small peaks sitting on a high background or less significant shoulder peaks. Furthermore, there are the advantages of the saving of deadtime in Wilkinson ADCs, of space on the storage disk, and of time for the analysis of the spectrum.

3.4.4. Backscattering bumps

It can be seen from Figure 4 that all energies of photons that initial energies over ca. 500 keV and which are scattered under an angle of ca. 180 degrees (backscattering) lie around about the same energy. This clustering of the backscattering energies, irrespectively of the initial energy of the photon, is the reason for the bumpy structure found around 220 keV, most prominently in spectra measured with shielded detectors. If the initial energies of the photons decrease considerably below 511 keV, the backscattered photons will of course be found at even lower energies, but the energy gap between the initial and the backscattered photons will become smaller and the backscattering bumps come closer to the full-energy peak. Thus, the backscattering bumps from the 79.8 and 81.0 keV peaks of ^{133}Ba are still well separated from the peaks whereas those of the 59.5 keV peak of ^{241}Am already merge with the full energy peak (Figure 5). Any analysis procedure will therefore have to consider this merging at lower energies and the sums of backscattering and full-energy peaks must be analyzed as sitting on one unique baseline.

3.4.5. The resolution (FWHM) function

When photons interact with the detector material most of the deposited energy goes into thermal vibrations of the crystal lattice. Only a small, constant fraction of the deposited energy is used to promote electrons from their ground states into the conduction band. The fraction used for this promotion process is defined by the Fano factor. This lifting of electrons into the conduction band is a statistical process, i.e. the number of electrons made available when an amount of energy (E) is deposited in the detector will have an average value N(E) with a variance of the order SQRT[N(E)]. In addition to this intrinsic variance of the signal one will then have further broadening through the noise of the linear electronics and the resulting resolution function should theoretically be described by a function of the form

$$\mathrm{FWHM}(E) = A + B * \mathrm{SQRT}(E) \tag{1a}$$

where FWHM is the full width at half maximum (in keV) of a full-energy peak from photons with an average energy E (in keV), and A and B are polynomial coefficients to be adjusted for each individual detector. In a spectrum, however, the experimentally available quantities are channel numbers and the number of counts per channel and thus the experimentally determined resolution function should be written as

$$\mathrm{FWHM}(\mathrm{channel}) = A + B * \mathrm{SQRT}(\mathrm{channel}) \tag{1b}$$

When one determines the resolution functions of Ge(Li) and HPGe detectors one will find, that there is rarely ever this theoretically expected linear dependence of FWHM with SQRT(channel), but that at high energies the FWHM is often significantly larger than expected. The reason for this enhanced FWHM lies in secondary effects such as charge density, impurity losses, recombination etc. which are more prominent when many electrons are promoted into the conduction band. A practical proposal for the spectrometrist therefore is that one should use a second order polynomial as a function of channel number

$$\mathrm{FWHM}(\mathrm{channel}) = A + B * \mathrm{channel} + C * \mathrm{channel}^2 \tag{2}$$

where A, B, and C are polynomial coefficients to be adjusted. This functional description has proven to fit all resolution functions well which the author has measured from a large variety of different p- and n-type germanium detectors. In most cases the coefficient C has a very small negative value.

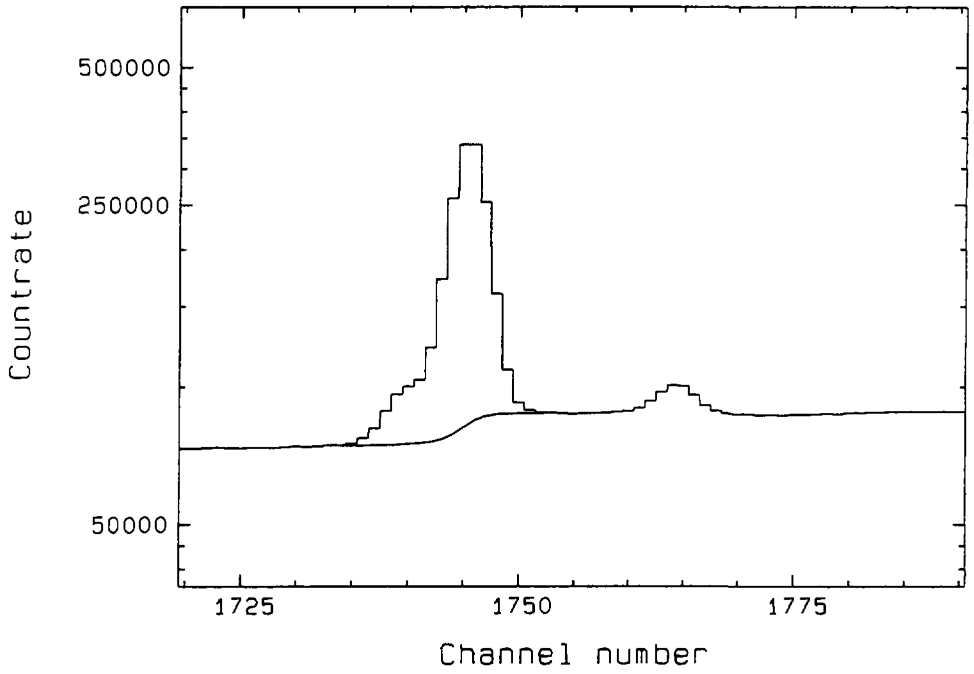

FIG. 2. Baseline distribution under an escape peak, which has a low-energy shoulder peak.

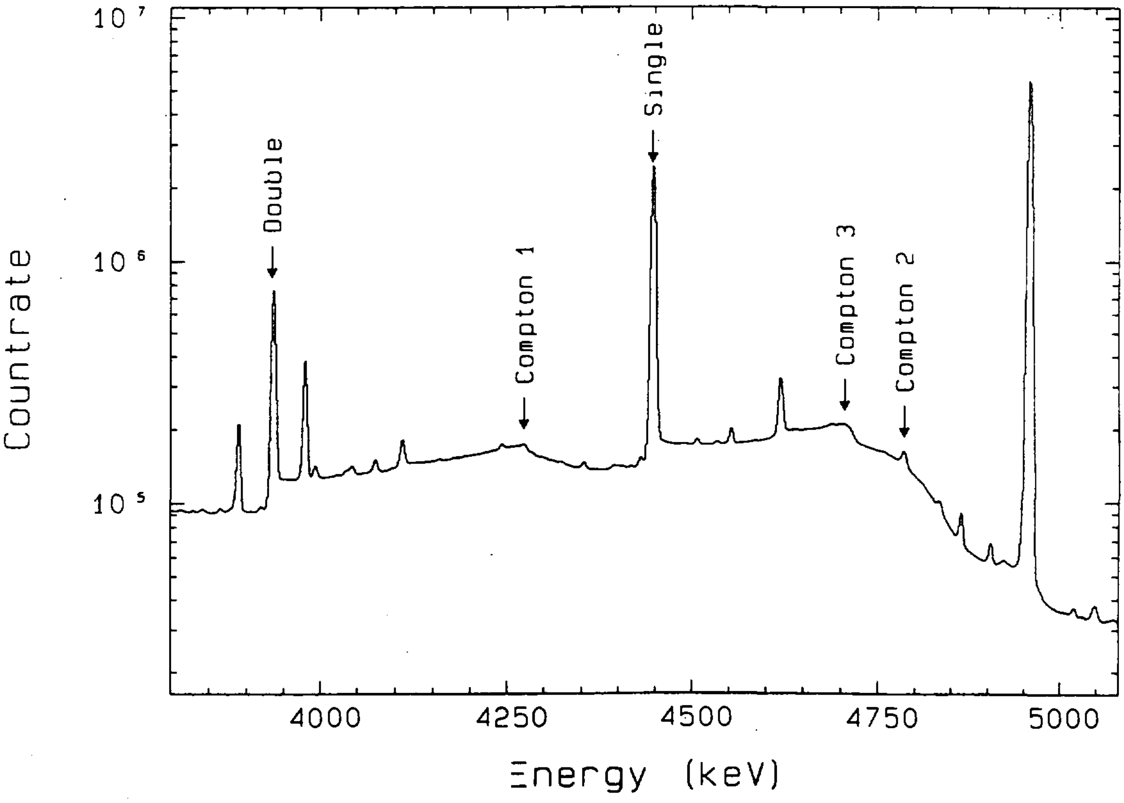

FIG. 3. A high-energy main peak and its escape peaks together with the Compton edges.

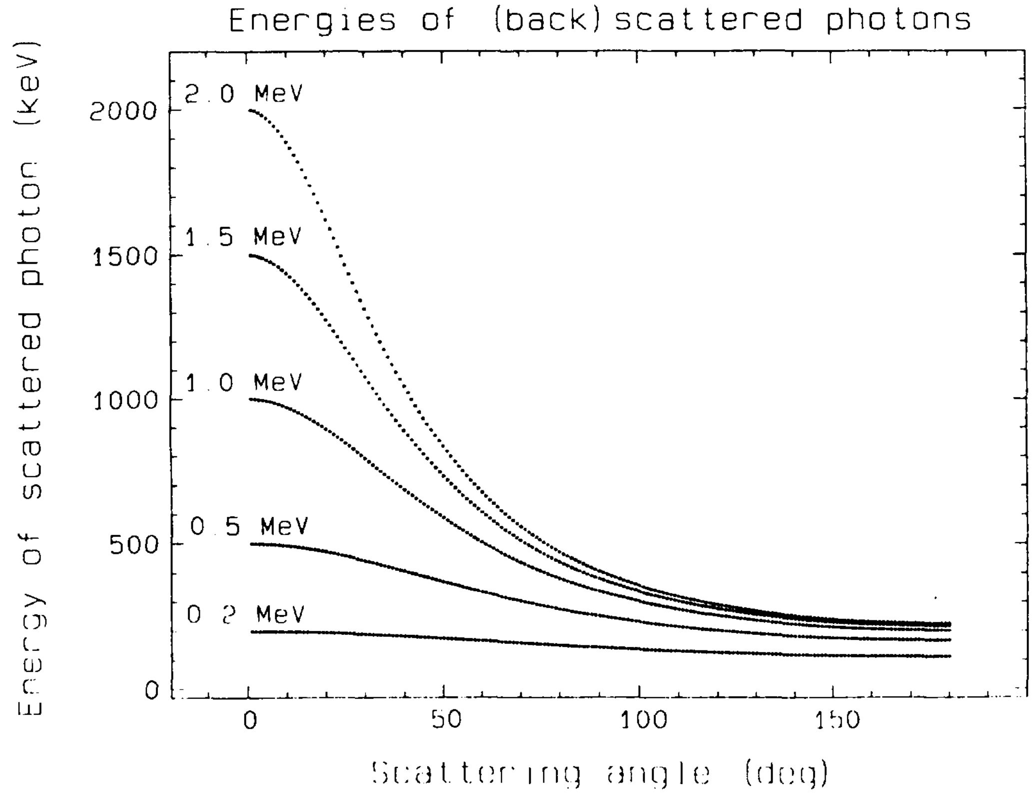

FIG. 4. Energy distributions as a function of scattering angle for photons of different primary energies.

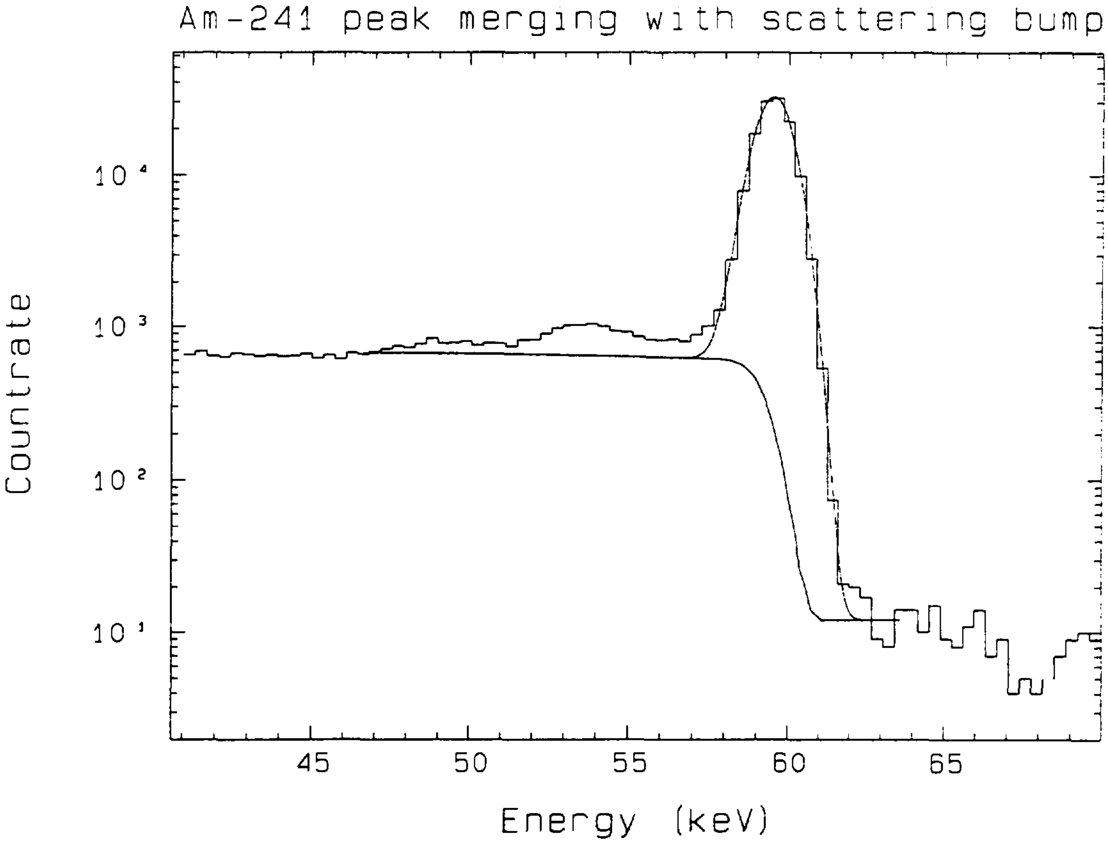

FIG. 5. The backscattering bump merges with the main peak at around 60 keV.

3.4.6. True coincidence summing

Whenever a decay process through negatron-, positron-, alpha decay or fission occurs, the electron shells of the daughter nucleus will be significantly distorted and X rays are emitted. In addition, the decay may leave the daughter in a short-lived excited state which then decays by the emission of a gamma ray or a cascade of gamma rays. Most of both these photon emission processes proceed on a time scale, which is too short to be discriminated by a semiconductor detector. Thus, if several photons originating from the same single decay process enter the detector the sum of the deposited energies will be measured. This is called a "true coincidence" and complicates quantitative analyses. Assuming that the emission of the photons reveals no angular correlation, i.e. gamma rays and X rays are emitted isotropically without any angular relation between one and the other, the probability that a second event is registered in coincidence with the first one is defined by the total efficiency for the second event. The total efficiency (as opposed to the full-energy peak efficiency) is the probability, that any that amount of energy of a photon is deposited in the detector. It considers all processes of energy deposition such as full-energy peak, Compton events, backscattering etc. The total efficiency is normally very much higher than the full-energy peak efficiency (for ^{137}Cs it may be 5 times larger) and as a consequence true coincidence summing may be very disturbing when the sample sits in close geometry and the efficiency is large. Every event assayed in coincidence with a full-energy event will "remove" the count from the full-energy peak and place it somewhere else (called "summing out"). When the coincident count is also a full-energy event the signal will be counted as a sum-peak event which may be another full-energy peak from the decay cascade (called "summing in"). At any rate, true coincidences remove counts from their proper locations in the spectrum. It will then be difficult to calculate activities because the full-energy peak efficiency function is no longer valid. One can calculate and correct for true coincidence effects [e.g. 2], but these calculations are very tedious because they need quite a bit of sophisticated experimental and tabular data input. The calculations need two sets of input data:

- One has to supply the total efficiency as a function of the primary photon energy. This data set must be measured once for each geometry in which samples are to be assayed. In order to determine the total efficiency function one should have a set of nuclides, which emit photons of only one single energy. The energies of the nuclides should cover the range from below 20 keV up to the highest energies to be found in the real samples. This set of nuclides is very difficult to find in most laboratories, which have no intense neutron source available.

- One has to supply the complete decay scheme for each nuclide where true coincidence corrections are to be applied.

  In some typical cases such as ^{60}Co, the amount of true coincidence losses from the full-energy peak efficiency function may be known and the correction applied accordingly. For most nuclides, however, the full-blown calculation of true coincidence correction factors is necessary, or these corrections should be avoided altogether. There are some practical solutions of how to deal with true coincidences in gamma ray spectra:

- The easiest way to avoid coincidence losses is to measure spectra at a large distance between the detector and source, so that the efficiency is small and true coincidences are negligible.

- In many applications it may not be necessary to quantify all lines from all nuclides contained in the sample. All lines or nuclides for which true coincidence losses to the count rates are known to be significant may be ignored.
- A "private" nuclides library may be created in which the apparent abundance of the gamma ray transitions for the nuclides are stored, i.e. the abundance as measured in a specific geometry by a specific detector. These "apparent" abundance have to be determined experimentally with each user's equipment and the user has to have different libraries for each specific setup. This is equivalent to needing different efficiency functions for each specific setup.

The latter method is definitely very tedious and time consuming but in many routine applications it has proven to be a very reliable and straightforward way to cope with the problem of true coincidence losses. Due to the fact that coincident X rays are encountered with almost every nuclear decay process it is especially important to apply coincidence loss corrections when n-type germanium detectors are used which measure these X rays with high efficiency. In fact, a practical recommendation is to avoid using n-type detectors for quantitative nuclide analyses.

3.4.7. Full-energy peak efficiency

There are several approaches to describing the full-energy efficiency as a function of the photon energy. In the most primitive approximation a linear function in a double logarithmic display (log(efficiency) vs. log(energy)) is used above ca. 200 keV and the absorption losses at lower energy are ignored. More sophisticated approximations use several polynomials which each describe a fraction of the efficiency function and which match smoothly at certain transition points. In most modern programs, however, the complete full-energy efficiency function is described by one unique function that is either a high-power polynomial [5] or the intrinsic function for p-type detectors [7]. For all n-type detectors as well as for those p-type units where one encounters a "dip" in the efficiency function around 600 keV, the polynomial representation seems to be the best suited description. When one uses the polynomial description one has to make sure that the whole energy range to be assayed in the sample spectra is also covered by as many as possible efficiency data calibration points. A high number of calibration points is essential because high-power polynomials will at any rate "bend" to match the data points but the functional trend in between the points may be rather arbitrary. Using 8 well defined data points and a fifth power polynomial we have found discrepancies between the fitted polynomial and the correct shape of the function of up to 8 percent of the correct value [7]. If one can supply very many data points over the whole energy range any such large deviation from the correct curve in regions between the points will be avoided. Furthermore, one may never apply a fitted efficiency function outside the range of energies which is confirmed through data points (extrapolation is forbidden!!).

It is a general rule that the efficiency function must be determined for each individual set-up in which samples shall later be assayed. The term "set-up" in this context means:

- One specific detector,
- One specific relative position between the source and the detector, (Strictly speaking, this refers also to the position of a source around the detector endcap. One may find a different efficiency when one measures a point source at the same relative height but 5 cm right or 5 cm left of the cap.)

- One specific sample geometry, (i.e. the shape and consistency of the actual sample which contains the activity. For example, if one uses always the same Marinelli beaker but its filling height is different in various measurements, or it is filled once with a fluid and in the next measurement with lumpy stone granules, then the sample geometry is not the same.)
- One specific sample composition.

If any of the first three of the above four conditions is changed one has to measure a new full-energy peak efficiency function. Modifications in the sample matrix composition will require additional corrections for self-absorption (see below) which could also be handled with a new efficiency function. It takes quite some effort to arrive at efficiency data points, which are as well defined as in the example function from Figure 6. A large number of well-calibrated expensive sources - some of which are very short lived - is needed to fill the energy range from below 50 keV up to almost 2000 keV. The sources themselves must be produced having exactly the same geometric distribution of the activity and they must be placed at exactly the same position. In many cases it is possible to produce one liquid mixed nuclide source from calibrated activity solutions, part of which is then poured into the desired geometry. The calibration spectrum should be counted for a sufficient amount of live time that all peaks to be analysed for the efficiency function have an area of at least 100 000 counts. The spectrum should be analysed with a real spectrum analysis program (most procedures implemented in the MCA emulation programmes are not suited because they apply too many approximations) and the peak areas must be corrected for true coincidence summing. If one considers the many complications and the amount of work involved to arrive at a good efficiency function, one might be tempted to scale that function linearly in order to correct for a different distance between the sample and the detector. Such linear scaling of efficiency functions (e.g. by $1/r^2$), however, must be avoided, as the efficiency functions measured at different distances do **not** run parallel. The *slopes* of the rising and the falling branches of efficiency functions will change when the source is placed at different distances. The amount of this change of slope is dependent on the type of the detector as well as the size and shape of the crystal. We have not yet been able to quantify the distance-scaling of efficiency functions in a systematic way.

3.4.8. Self-absorption corrections

Whenever a source of gamma rays has a finite thickness the gammas will interact with the matrix of the source and be partly absorbed. There may be additional absorption between the emission and registration of the photon such as interactions in the container, in the air between source and detector, in the detector endcap, and in the dead layer on the detector. The effects of absorption are most prominent when the source is thick, like in a Marinelli beaker, and when it consists of material with high atomic numbers. There are many attempts to calculate or approximate corrections for photon absorption effects [e.g. 1, 2, 4, 9, 11, 13, 14] some of which yield quite consistent and acceptable results. However, it seems to be the simplest, most precise and best method to determine experimentally several self-absorption corrected efficiency functions for each typical source geometry [10] and to use the best suited one. As an example several normalized efficiency functions are shown in Figure 7 which were measured in a Marinelli-beaker geometry from calibration nuclides mixed into sources of different matrix compositions.

The matrix densities of the sources were in the range of $0.42\ \mathrm{g/cm^3} \leq \rho \leq 1.60\ \mathrm{g/cm^3}$ and the atomic numbers Z of the matrix elements were in the range of $1 \leq Z \leq 26$. The functions in the figure are the ratios of the efficiency functions measured in a specific matrix divided by the efficiency function measured in a polymerized ethylene (brandname: Lupolen). From the varying shapes of the functions and even the crossing of different functions it is clear that any density- or Z-dependent scaling of a reference efficiency function will not yield a reliable method of simultaneous absorption correction. One can, however, use the appropriate efficiency function out of a set of measured data in order to correct for the absorption in an actual sample. The function to be used is easily found through an absorption measurement at two significantly different energies [12, 10]. The efficiency function from that sample matrix is then to be used, in which the ratio of the attenuation coefficients for the two energies is nearest to the ratio found in the actual sample.

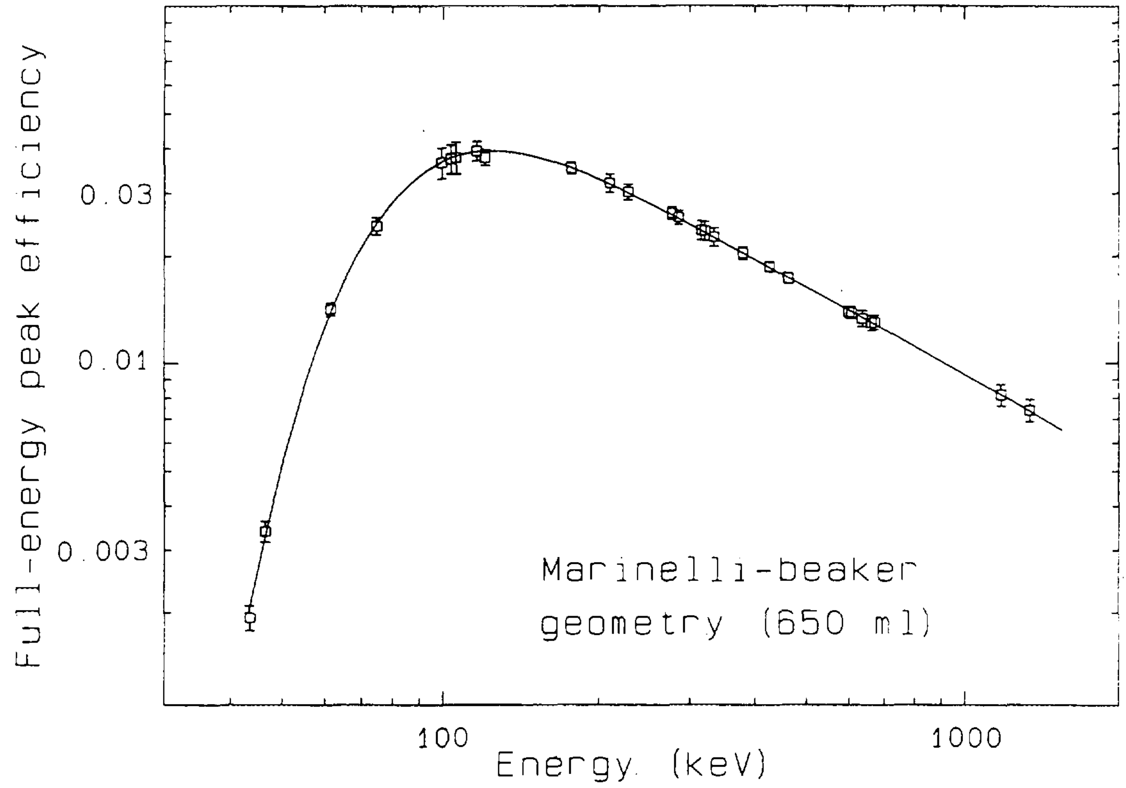

FIG. 6. Full-energy peak efficiency function for a 650 ml Marinelli-beaker geometry.

3.4.9. Uncertainty calculation

It is well known that the minimum 1-STD statistical uncertainty of a large number of counts (N) is approximated by SQRT(N). This simple formula may be applied if all counts originate from the same process and they are generated and treated in the same way. However, it does not apply when the area uncertainty for a peak in a gamma ray spectrum is to be defined since

- the peaks in a spectrum always sit on top of a baseline distribution, there may be one or several other peaks whose area overlaps with the one peak under inspection, and
- the definition of a region-of-interest (ROI) to be analysed is arbitrary.

These additional constraints are to be considered in the analysis of the spectrum and their contributions to the overall uncertainties of the peak areas must be reflected in the area of uncertainty. Therefore one may not define the area uncertainty as x-times SQRT(area) where x is a convenient multiplier. The uncertainty must consider the deviations in each channel of the fitted from the measured counts, the contributions of the baseline counts, the partition of count rates to different peak areas, and also the possibility of missing or extra peaks in the analysis of a ROI. Unfortunately the sum of all these and possibly other sources of error leads to total uncertainties which are much larger than users wish them to be. In most programs for the analysis of gamma ray spectra, there is no such very detailed estimate of uncertainties but rather, a few simplifying assumptions are made. These are:

- the border channels of a ROI are correctly defined,
- all peaks in a ROI are consistently determined, there are no missing nor extra peaks,
- the peak shape used in the fitting process is physically correct,
- the functional description of the baseline is physically correct,
- all peaks in one ROI have identical peak shape parameters.

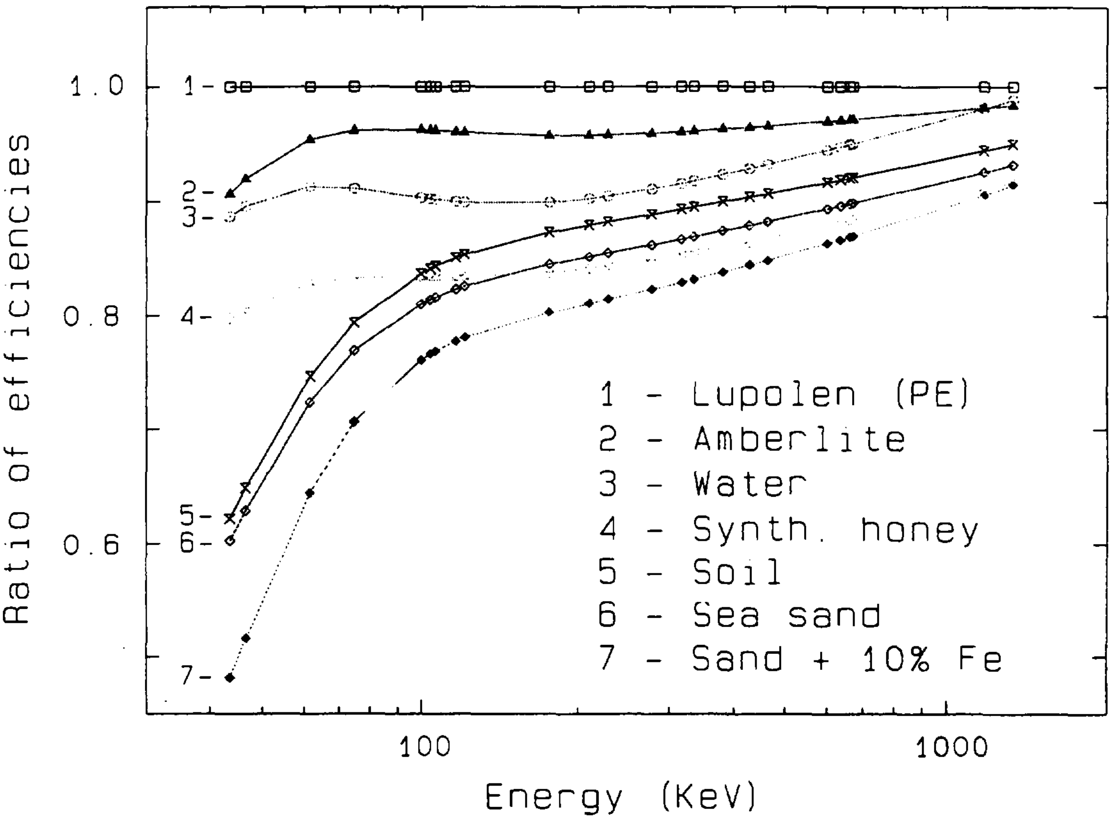

FIG. 7. Ratios of self-absorption corrected efficiency functions measured in the same geometry but using different matrix materials. The lines are to guide the eye.

Provided that these assumptions are justified and that the least-squares fitting process has really found the minimum deviation in the multiparameter space, then the area of each peak can be calculated as the integral over the peak function from -∞ to +∞. In many cases this integral can be solved in a closed form and the area is written as a function

$$AREA = f(a,b,c,...) \quad (3)$$

where a, b, c, ... are functional parameters whose numerical values are either known or approximated in the fitting process. The 1-STD uncertainty of the area then is calculated as

$$\Delta AREA^2 = (\delta f/\delta a)^2 \bullet \Delta a^2 + (\delta f/\delta b)^2 \bullet \Delta b^2 + \ldots.. \qquad (4)$$

where Δa, Δb, ... are the 1-STD uncertainties of the parameters. These are taken from the estimated covariance matrix [C] of the standard uncertainties in the fitted parameters α: $[C]=[\alpha]^{-1}$ (if a nonlinear matrix algorithm is applied) or from the properly weighted residua (if a nonlinear, non-matrix algorithm is used). Again, these methods to define the uncertainties of fitted parameters are only justified if the resulting uncertainties in repeat analyses are normally distributed. In spectrometric analyses this will normally be the case if the above five assumptions are justified, i.e. if the analysis was "*correctly*" made.

The final result from most spectrometric analyses is the quantitative definition of activities from nuclides in the sample. This quantification requires calibration operations that are based on fitted experimental data and therefore also have uncertainties, such as the efficiency calibration or the self-absorption corrections. The uncertainties coming from these calibration procedures must be considered according to the statistical law of uncertainty propagation (equation 4) when the total uncertainty is calculated. Uncertainty contributions from these calibration procedures can be very large, as shown in Figure 8 where the example of the well-determined efficiency function from Figure 6 is employed. Figure 8 shows that the ratios between the experimental data points and the fitted function, i.e. the solid line at unity marks the fitted function. The enveloping functions above and below the data points are the limits of the 1-STD uncertainty of the fitted function as calculated from equation 4. Although this efficiency function is very well determined through data points and it is well fitted, there are very large contributions up to 10% to the overall uncertainty, especially at low energies. These contributions must be considered when peak areas are corrected for efficiency and activities are calculated. Thus, in the above example a nuclide which is assayed through a low-energy line alone (such as e.g. ^{210}Pb via the 46.5 keV line) cannot be determined to better than ±10 percent.

3.4.10. Spectrum formats

Most modern multichannel analyzers allow the storing of measured spectra on permanent mass storage devices such as the hard disk, floppy disk or tape. This very useful feature, however, suffers from the fact that there is no agreed standard of how these spectra should be formatted. It is a bad habit that every supplier of spectrometric hardware has his own spectrum format, which is by principle incompatible with every other format. As an additional complication one may find several different, internally incompatible formats in different hardware from the same supplier, and as an extra surprise, one supplier even had a total of three different formats used in the same hardware. The differences are found in the length, contents and structure of header and trailer blocks as well as in the representation of the spectral data. The latter point is especially annoying because there is good emulation software, which allows the storing of data with 32 bits per channel or a poor system that provides only 20 bits for each channel. On 20 bits, however, one can only store data with a maximum of 1 048 575 counts in one channel, which is definitely not sufficient for any spectrometric purpose. Counting only the more abundant MCAs, there are more than 20 different spectrum formats on the market, which are completely incompatible with each other. This situation is most unsatisfactory, and users ought to make frequent complaints in order to pressure producers to agree on one standardized spectrum format.

As a starting point the author would propose that an ASCII header and spectrum format be used which resembles the structure employed in the current QXAS file format of the IAEA programs. The format is set up in a keyword-controlled structure with a limited number of pre-defined keywords and any number of user-defined keywords. Thus, there is no limitation for each hardware supplier to implement his own ideas and options. As an example, a keyword controlled spectrum format might look like the following:

```
$channels$8192
$realtime$1007
$livetime$1000
$deadtime%$0.4
$startgrab$21 aug 94 16:30:04
$endgrab$7 oct 94  09:04:51
$startcount$12 oct 94  08:01:09
$mass-g$17.941
$begin_data$
0 0 0 0 0 0 0 0
0 0 0 0 0 0 0 9
14 36 102 367 1009 2047 2081 2107
...
...
$end_data$
```

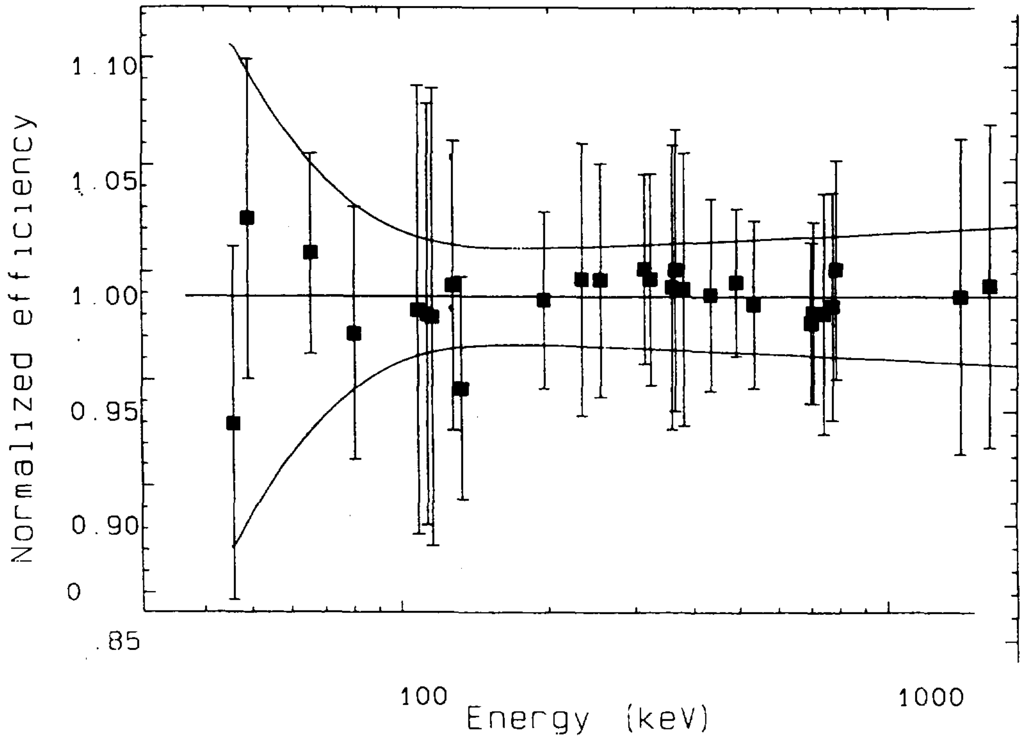

FIG. 8. Efficiency data points from the function from Figure 6, normalized to the fitted function. The enveloping lines show the 1-STD uncertainty band of the fit.

In the above example the keywords are enclosed in "$" signs for easy decoding. A similar proposal of a keyword-controlled ASCII spectrum format has been made before [8] and it could also be pursued. It is an unnecessary waste of user's time and programmer's brain to have to handle so many complicated, poorly documented and inaccessible spectrum formats. The argument that ASCII formats are too space consuming and expensive for practical purposes is no longer valid, because in today's computers the price of mass storage space is below 30 cents for 1 MB of storage capacity and it will drop further.

3.5. CONCLUSIONS

The techniques of measurement and evaluation of high-resolution gamma ray spectra have been developed and refined since over 20 years and are generally good and reliable now. However, there are still several problems, inconsistencies and simplifications that require solutions and improvement. In the present paper I have pointed out and proposed solutions for the most frequently occurring hardware problems as well as some of the areas in which existing software may need modification. Of these problems, there are two important topics for which there are not yet practical, easy-to-use solutions. They relate to the quantification of activities in samples and involve the true coincidence summing and the self-absorption corrections. Ways to approximate these two correction functions have been discussed, but it would certainly be desirable that basic solutions for these problems be found.

REFERENCES TO SECTION 3

[1] BIRATTARI, C., SALOMONE, A., Efficiency evaluation of gamma ray solid-state detectors, Nucl. Instr. Meth. **174** (1980) 391.

[2] DEBERTIN, K., SCHÖTZIG, U., Coincidence summing corrections in Ge(Li)-spectrometry at low source-to-detector distances, Nucl. Instr. Meth. **158** (1979) 471.

[3] DEBERTIN, K., REN, J., Measurement of the activity of radioactive samples in Marinelli beakers, Nucl. Instr. Meth. in Phys. Res. **A278** (1989) 541.

[4] DRYAK, P., KOVAR, K., PLCHOVA, L., SURAN, J., Corrections for the Marinelli geometry, J. Radioanal. Nucl. Chem. Lett. **135** (1989) 281.

[5] GRAY, P.W., AHMAD, A., Linear classes of Ge(Li) detector efficiency functions, Nucl. Instr. Meth. in Phys. Res. **A237** (1985) 577.

[6] HELMER, R.G., LEE, M.A., Analytical functions for fitting peaks from Ge semiconductor detectors, Nucl. Instr. Meth. **178** (1980) 499.

[7] JÄCKEL, B., WESTMEIER, W., PATZELT, P., On the photopeak efficiency of germanium gamma-ray detectors, Nucl. Instr. Meth. in Phys. Res. **A261** (1987) 543.

[8] MANATT, D.R., CARLSON, J.B., GUNNINK, R., RUTHER, W.D., BUCKLEY, W.M., A proposed ASCII transfer spectral data file format, LLNL-Report, Livermore (Feb. 1990).

[9] NAKAMURA, T., SUZUKI, T., Monte Carlo calculation of peak efficiencies of Ge(Li) and pure Ge detectors to voluminal sources and comparison with environmental radioactivity measurement, Nucl. Instr. Meth. **205** (1983) 211.

[10] PÜTZ, M., Gammaspektrometrische Bestimmung Umweltrelevanter Radionuklide In Optimierter Marinelli-Geometrie, Thesis (1994) Philipps-Universität Marburg.

[11] SANDERSON, C., A Procedure For Incorporating Gamma-Ray Attenuation Corrections Into Germanium Efficiency Calibrations (1995) this report.

[12] SATOH, K., OHASHI, N., HIGUCHI, H., NOGUCHI, M., Determination of attenuation coefficient for self-absorption correction in routine gamma ray spectrometry of environmental bulk sample, J. Radioanal. Nucl. Chem. **84** (1984) 431.

[13] SIMA, O., Monte Carlo simulation versus semiempirical calculation of autoabsorption factors for semiconductor detector calibration in complex geometries, Progr. Nucl. En. **24** (1990) 327.

[14] SIMA, O., Photon attenuation for samples in Marinelli beaker geometry: An analytical computation, Health Phys. **62 5** (1992) 445.

[15] WESTMEIER, W., Background subtraction in Ge(Li) gamma-ray spectra, Nucl. Instr. Meth. **180** (1981) 205.

[16] WESTMEIER W., The fitting of solid state detector spectra, Nucl. Instr. Meth. in Phys. Res. **A242** (1986) 437.