A Paradigm Shift in Nuclear Spectrum Analysis

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***Abstract* - An overview of the latest developments in quantitative spectrometry software is presented. New strategies and algorithms introduced are characterized by buzzwords "Physics, no numerology", "Fuzzy logic" and "Repeated analyses". With the implementation of these new ideas one arrives at software capabilities that were unreachable before and which are now realized in the GAMMA-W, SODIGAM and ALPS packages.**

I. The Development of Nuclear Spectrometry

The advent of drifted semi-conductor (germanium or silicon) detectors in the early sixties opened a completely new era of quantitative nuclear radiation spectrometry. Former scintillation detectors having reasonable spectrometric properties such as NaI(Tl) were rapidly replaced by Ge(Li) and Si(Li) detectors cooled with liquid nitrogen, and by diffused junction or surface barrier detectors for (α-)particle spectrometry in vacuum at room temperature. The next major step in the development of detector technology came with refined techniques for hyper-pure germanium material production in the early 80s that allowed the making of un-drifted HPGe detectors for penetrating radiation. At about the same time a novel technique of ion implantation allowed fabrication of particle detectors without the disturbing dead layer on the surface, leading to an improvement in resolution, and to cleanable (washable) detector surface.

Multichannel-analyzers (MCAs) used for NaI(Tl) scintillation spectrometry typically measure spectra with up to 1024 channels, where peaks are sufficiently well resolved for quantitative analysis. High resolution Ge(Li) and HPGe spectrometry, however, requires at least 4096 channels spectrum length and modern spectrometers provide 8k or even longer spectra. The technology of MCA production advanced rapidly in the 50s after the development of semiconductors. Whereas the first MCAs were bulky and very heavy because they were operating with vacuum tubes and relay counters, the invention of transistors immediately transformed the MCA into a nice little stand-alone unit that could even be carried by just one experimentalist. With the advent of personal computers in 1981 and improved high-integration electronics, MCA technology migrated into the PC and plug-in MCA cards were developed that even contained high voltage power supply for HPGe detectors, spectroscopy amplifier, gain stabilizer and very fast fixed dead-time ADC. This integration of hardware into the PC has now been abandoned because of excessive development cost prompted by continuous change of the PC bus structure. Modern spectrometry hardware is again designed as stand-alone or NIM unit that links into the computer via USB, RJ-45 network or wireless interface.

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Quantitative spectrum analysis was made in the beginning via summing of counts and linear baseline subtraction for singlet peaks. Multiplets were analyzed through graphical peak separation and gravimetric integration. When mainframe computer centres were set up in universities, scientists began to develop a myriad of spectrum analysis software, only a few of which were actually very powerful and successful. The ANS topical conference on "Computers in activation analysis and gamma-ray spectroscopy" in 1978 presented a large number of software available at that time [1]. Similar to MCA development, the advent of PCs changed the scene, and it brought another torrent of spectrometry software. However, the number of good software yielding reliable results did not increase significantly. Many programs assumed simplified Gaussian peak-shapes and linear baselines, whereas others excelled with over-complicated numerology without any relation to physics. Good software has been commercially available from various vendors since the 80s. It is interesting to note that several sophisticated programs for the analysis of high-resolution gamma-ray spectra are available but the selection of programs for the analysis of scintillator spectra is very meagre. This deficiency is particularly strange, because scintillation spectrometry is a frequently used method in many industrial and medical applications. All scintillator programs except one fit approximated shapes to the peaks and very few calculate the correct shape of the baseline. Similarly serious deficiencies are encountered in almost all programmes on the market for the analysis of high- or low-resolution alpha-particle spectra.

The following sections give an overview of modern spectrum analysis principles and strategies.

II. Spectrum Analysis Software

After 1981, the development of software for the analysis of energy-dispersive spectra from nuclear decay has more or less followed the technical development of personal computers, and major emphasis has been on user's request for simplified and intuitive handling. The latest commercial or freeware programs on the market such as for example the GammaVision package with its many application-specific variants [2], the Genie-2000 package [3], the HyperLab 2005 and 2009 packages [4] which are copies of the former Hypermet program [5], FPGAS [6] which is a copy of Sampo90 [7], InterWinner [8], SpectLab [9] or DEIMOS32 [10] are basically just developments for new operating system environments. None of the above mentioned and other software for quantitative nuclear spectrometry provides new algorithms or strategies for improved spectrum analysis. However, such improvement will be presented below as it is realized in our GAMMA-W, SODIGAM and ALPS packages

for quantitative high-precision analysis of gamma-ray and alpha particle spectra.

All programs can analyse big singlet peaks correctly and with an acceptable level of accuracy, though it may seem that uncertainties of least-squares fitted parameters and of peak-areas are significantly underestimated by some programs.

A few general statements are:

- All programs run under WINDOWS®
- All commercial programs have software protection units
- All reasonable programs can read various spectrum formats
- All reasonable programs can make reliable automatic spectrum analyses
- All reasonable programs provide a software update service
- All reasonable programs provide on-line help and hotlines
- All reasonable programs provide spectrum oriented analyses but not peak-list oriented analysis.

Major inconsistencies are found in many programs where purely mathematical models are applied for the analysis of spectrometric properties and used to quantify peaks in the spectra (i.e. the numerology approach). Typical examples of inappropriate modelling are the linear or parabolic baseline under a peak, the purely Gaussian peak-shape, the square-root dependence of FWHM on energy, or the high-order polynomial description of the shape of a full energy peak efficiency function for γ-rays. Correct spectrum analyses can only be made when physics-oriented descriptions of relevant spectrometric properties are found and applied in spectrum analysis. It is not meaningful to assume a straight or parabolic line or another simple mathematical model for the shape of the baseline under a peak or multiplet. There is only one physical principle how the baseline is produced in the interaction of gamma-rays in the detector and there is only one shape of the averaged external background.

As an example, we will deduce the shape of the baseline under a peak. It is clear that inclusion of counts that lie under the baseline of a peak will yield an excessively large peak-area and thus one has to separate the background counts prior to peak or multiplet analysis. This problem was first tackled by Pratt [11] and a review of commonly used nonlinear baseline shapes was presented by Helmer and Lee [12] A graphical display of some of Helmer's examples is shown in Figure 1 where the peak-position is in channel zero. Simple examples such as a straight line are omitted in the figure. In various papers it was later shown [13-15] how one can accurately calculate the correct shape of the baseline under a peak or multiplet exclusively from the numbers of counts in the channels in a region under consideration. The method works without any model assumptions or arbitrary peak-shape description. The procedure used is based solely on the physics-oriented finding that the background in the low-energy vicinity of a peak is constant, and it is smooth and continuous on the high-energy side, as shown in Figure 2. Moreover, there is a different height of the background before and after the peak, and the peak itself is almost a delta function. The method for baseline definition as described in refs. 13-15 is based solely on these few simple and apparent facts. It allows one to calculate the baseline and subtract background counts from the spectral region to be analysed prior to the peak fitting procedure. There is no need to know the numbers, locations and shapes of peaks in the region where the background is to be determined. The difference between simple (mathematical) baseline functions and the correctly calculated one may amount to less than 0.3% of the peak-area of a large singlet peak. The importance and validity of the method proves itself when it comes to the deconvolution of multiplets, especially where a small peak sits in the shoulder of another big one. Only when the physically correct baseline is calculated and subtracted from the multiplet, will the fitted area of the small shoulder-peak match the correct value.

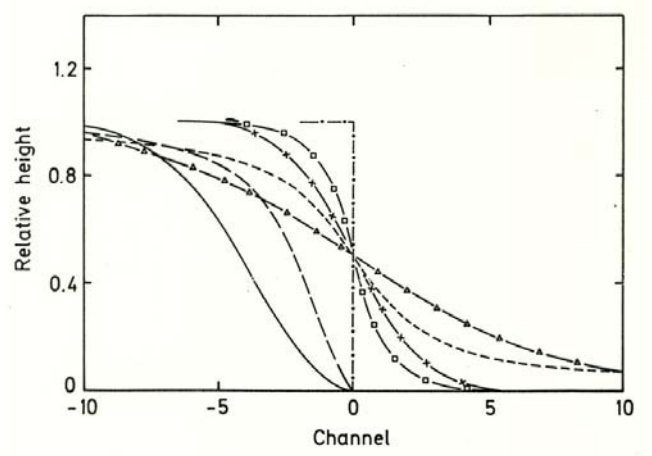

Figure 1. Compilation of baseline-functions under a gamma-ray peak (after Helmer and Lee [12])

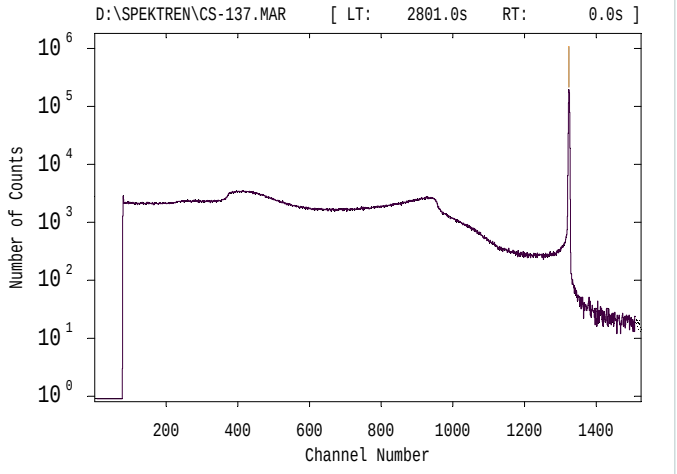

Figure 2. Spectrum with one singlet full-energy peak

Another example of inappropriate numerology in peak analysis is the choice of purely mathematical descriptions of peak-shapes for the fitting process. Most programs adopt the physically supported assumption that the main contribution to a gamma-ray peak is a Gaussian distribution. A variety of model shapes are then assumed in order to describe the low-energy and sometimes also high-energy tailing of the peak. Such numerical approximations for peak-shapes without any reference to nature are not an acceptable solution. The only acceptable procedure is to find out what the peak-shape actually looks like and then utilize that physics- or experience-

oriented description in the spectrum analysis program. In this context, it should be noted that peak-shapes encountered for the old Ge(Li) detectors are different from shapes of modern HPGe detectors, and that the shapes found in very large HPGe detectors again may require different description.

A completely new strategy towards programming spectrum analysis has been introduced in recent years. The heart of the method lies in the fact that there are often various different ways in which a solution to a problem can be found. Some ways may be good for one situation but poor for another, some procedures are generally usable whereas others may be suitable for only very specific cases. The modern approach to handling this dilemma of "how to do it right?" is called FUZZY LOGIC where one:

- uses all available methods and algorithms
- assigns weights to the various results, based on the applicability of the respective method
- discards inapplicable methods
- calculates the weighted average of valid results.

The averaged result of properly weighted individual solutions is always better, more stable and more significant than any single result. Typical applications in a spectrometry program for Fuzzy Logic solutions are, to give only a few examples, the definition of regions in a spectrum that may contain peaks, the definition of the average height of the background before and after a peak or multiplet, the automatic generation of the resolution function, the assignment of nuclides to detected peaks, or the initial search for potential peaks and shoulder peaks.

A very difficult task is the definition of all peaks that must be fitted in a selected region of the spectrum. There are programs that deduce the list of possible peak candidates from the user-defined library. This method of library-oriented spectrum analysis, however, intrinsically has a serious deficiency. Nuclides that are missing in the library will never be detected and, often worse than that, missing peaks will lead to erroneous analysis of other peaks in the fitted region. The fully automatic analysis of a spectrum for determination of peak-positions and peak-areas basically requires a priori knowledge of all peaks that exist. As this premise is never fulfilled the program must analyse regions under the assumption that detected major peaks constitute all components. The residuum analysis of a fitted region may then reveal the presence of more peaks, or the analysis of statistical significance or peak proximity may render some of the fitted peaks as actually non-existent. A new fit of the same region but with a different set of potential peak candidates may yield an improved fit and finally, sometimes after three or four attempts to analyse a region, the statistically best possible solution will be found. Because of the very high computing power of modern PCs, such iterative region analysis is not a practical problem at all.

Examples of peak-fit results using modern commercially available programmes for spectrum analysis are displayed in Figures 3 to 5. A section from a HPGe spectrum taken from a U/Th source is shown in Figure 3 together with the course of the baseline and automatically fitted peaks. The sum-function of baseline plus peak contributions is also shown and it goes smoothly through the measured spectrum data shown as a histogram. As no peak-search algorithm will à priori find small peaks around channels 1372, 1380, 1390 and 1395, such complete automatic quantitative analysis of a region is only possible when consistent definition of peak FWHM with respect to neighbouring peaks or regions is controlled and when the region is subject to repeated analyses using various sets of possible peaks. Just to mention, the correct definition of uncertainties to measured numbers of counts in the spectrum (considering robust statistics) is an essential aid for finding missing peaks without an over-definition of spectrum components.

In Figure 4 an unusual alpha-particle spectrum having very poor statistical precision is shown which was taken with an ion-implanted detector from a thin layer of "powder sample" consisting of ground Pitchblende material. The average grain size of the powder was around 60-100 µm which is infinitely thick with respect to the range of alphas in material and therefore renders the material not suitable for high-resolution alpha spectrometry. Tails of peaks extend down to zero energy because alpha particles can be completely stopped within the thick sample. The maximum number of counts in one channel is around 60 counts, except in the low energy regime where beta particles and noise contribute to the spectrum.

Using the physics-oriented peak-shape that is expected for such sample material, i.e. correctly considering the energy-loss of α-particles between emission and registration, which

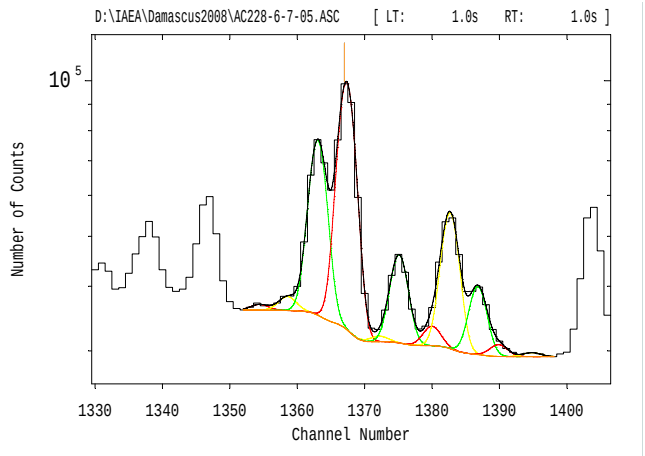

Figure 3. Fully automatic analysis of a region around 1001 keV in the γ-ray spectrum from a U/Th-containing source

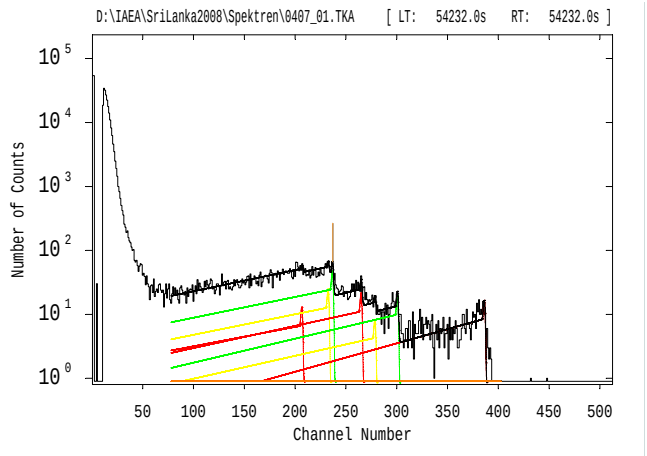

Figure 4. Analysis of an alpha spectrum taken from a sample of pitchblende powder containing ^{238}U and progeny

leads to a quite complicated mathematical description of the peak- and tailing-shape, one will find the fit presented in Figure 4 where all alpha-emitting nuclides from the ^{238}U decay chain are identified and consistently quantified. Resolution into various contributing peaks for one nuclide as well as high-precision analyses yielding peak-areas with very small uncertainty are of course impossible in low-resolution spectra taken from powder material. The striking advantage of the new possibility to analyse alpha-spectra taken from powder or filter samples is the very ease of sample preparation. There is no more need for chemical dissolving of the specimen, element separation and deposition of a very thin sample. One can rather grind the sample material with a ball-mill or other suitable mechanical device and measure a thin layer of the powdered sample, or take a pressed filter as is, make the measurement and quantify analytes with reasonable accuracy.

In Figure 5 a region out of a ^{166m}Ho spectrum is shown which was measured with a $LaBr_3$(Ce) scintillation detector. The indicated region was automatically defined and analysed without user-intervention. Using the correct peak-shape description for this type of detector material, proper FWHM description and calculation of the baseline according to the physical interaction of photons in that material the software can find and fit the region as indicated. Areas from all peaks except the 736.5 keV peak (0.382% intensity) around channel 495 agree well within ±1σ uncertainty with literature values.

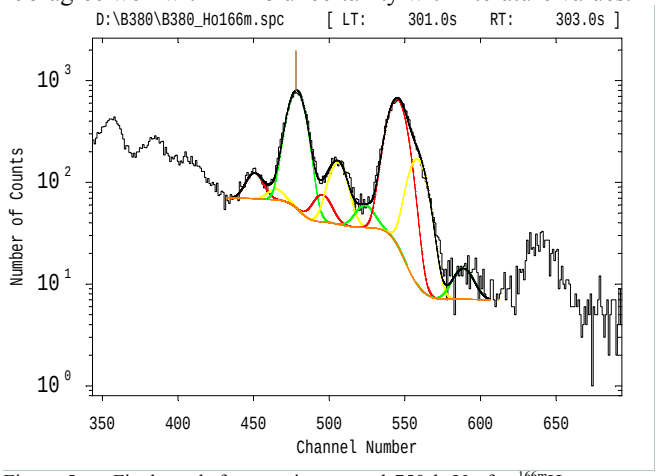

Figure 5. Final result for a region around 750 keV of a ^{166m}Ho spectrum which was measured with a 2"x2" B380 detector and automatically analysed

Only through the consistent combination of methods and strategies described above, namely physics-oriented modelling, Fuzzy Logic strategies in solution finding and re-iterative analyses of regions can modern programs provide correct and fast analyses of spectra and yield reliable results for peak-positions and peak-areas of all peaks that can be analysed from the spectrum. Actually, through combined use of these new ideas a complete paradigm shift in nuclear spectrometry has been achieved. The old "von Neumann" course of straightforward programming in spectrum analysis is no longer used, but it is rather replaced by "multi attempt" spectrum analysis strategies employing (admittedly complicated) models which describe spectrometric appearance very well and follow physical reality very closely.

A very thorough and demanding investigation and inter-comparison of commercial and freeware PC-based γ-ray spectrum analysis software has been made some time ago by the International Atomic Energy Agency [16]. Only three out of the twelve tested programmes were listed, which "with respect to resolving power as defined in this test and quality of area determination ....yielded the least bad results". Thus it is clear that good spectrometry programs were not easily found and new ideas and strategies were required.

III. Conclusions

The field of quantitative nuclear spectrometry is not dead. New developments of MCA hardware, the improvement of resolution in traditional NaI(Tl) crystals and especially the development of new scintillator materials such as $LaBr_3$(Ce) or $CeBr_3$ with significantly improved properties compared to NaI(Tl) has opened a completely new range of applications and possibilities. Many measurements can now be made with room-temperature scintillation detectors where the use of cooled HPGe detectors was once mandatory.

Modern spectrum analysis programs use improved algorithms; in particular the modelling of peak-shapes, baselines and other spectrometric properties is now based on physical correctness rather than on numerical simplicity. The use of Fuzzy logic improves many tasks in automatic spectrum analysis and it allows much better definition of reference points and dynamic variables. Multiple analyses of the same region using different reasonable sets of peak references are a good tool for successful result optimisation.

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