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ANALYSIS OF MULTIDIMENSIONAL SPECTRA

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Coincidence spectra obtained from arrays of radiation detectors can be analyzed without resorting to projecting lower dimensional spectra. The techniques for two-dimensional analysis (peak searches and peak fitting) are described, and extensions to higher dimensions are discussed. Even for two-dimensional analysis it is most efficient to compare predictions from a model, such as a scheme for radioactive decay, with the data by backcalculating the data from the model.

Nuclear and subnuclear processes are perhaps unique among quantum-level events in the opportunity to characterize single events–the decay of a radioactive atom or the interaction of an energetic particle. This opportunity extends to simultaneous measurements of more than one feature of single events, namely, the coincidence method.[1] The combination of many coincidence events leads to multidimensional spectra in the sense used in this paper, even if the events remain as a list rather than a histogram.

Detector arrays[2] have been developed to study deexcitation of nuclides produced in nuclear reactions. Current emphasis on nuclear rotational features at high angular momenta leads to highly segmented detectors in order to avoid excessive "double hits" from the large number of coincident gamma rays. In general, radioactive decay does not involve such large numbers of coincident emissions, but the availability of these arrays has led to the construction of decay schemes far too complicated to have been worked out in a reasonable time by measurements with pairs of detectors. For example, the spontaneous fission of ^{248}Cm has been studied[3] in EUROGAM 1, which was an array of 45 suppressed Ge detectors and 5 Ge detectors with good efficiency and resolution at x-ray energies. We can consider spontaneous fission to be a form, albeit extremely complicated, of radioactive decay.

Data obtained from these arrays contain an enormous amount of information, but extracting that information is a major challenge. Two-fold coincidences among 50 detectors involve 1225 two-dimensional spectra, each of which might require 100

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megabytes for storage as histograms. The number and sizes of spectra of three-fold coincidences are even more impressive, but three-fold coincidences are necessary for constructing complex schemes.[3] Of course the data are recorded as lists (of pulse heights, times, and detector IDs) which can be considerably shorter than the histograms they imply. In practice histograms are combined; if angular distributions are not being sought, all of the similar detectors can be normalized to the same calibration and events of a given fold can be combined into a single histogram.

How are the histograms analyzed? A portion of a 2D histogram is given in Fig. 1. Main features to note, in addition to the obvious peaks, are the ridges and the 2D continuum. The most common method for analyzing coincidence spectra is to set "gates" on one or more of the parameters and obtain lower-dimensioned spectra, which in turn are analyzed by familiar techniques (e.g., SAMPO,[4] GAMANAL,[5] and HYPERMET[6]). The logic of "gating" can be illustrated by its application to the reduction of a 2D peak to the net number of counts in the peak. Consider the diagram in the right side of Fig. 1. In this case the peak in each dimension comprises four channels, and two channels to either side of the peak can be used to correct for the continuum. The sequence is: start with one of the parameters, add the spectra in the peak channels, and subtract the spectra in the continuum channels (if such can be found). The process is continued with the second parameter, with counts being added and subtracted. The net effect is to equate the intensity (volume) of the peak to the sum of the counts in the shaded channels minus the counts in the unshaded channels. The 2×2 portions at the corners arise from the product of two subtractions and contribute to the noise (statistical errors) but not the signal. A better result would be obtained by subtracting one third of the sum of surrounding 48 channels from the 16 peak channels if, as in single parameter spectra, features were confined to peaks and continuum. However, without taking the ridges into consideration, the intersection of the ridges from non-coincident gamma rays would look like a peak.

The same statistical considerations apply when the gates are used to reduce the data to one-dimensional spectra, which are analyzed by the same peak-fitting programs that have been developed for simple one-dimensional spectra. These fitting programs are certainly more sophisticated than simple sums-and-differences, but neither the absolute values of the peak intensities nor the reported uncertainties in energies and intensities can be trusted if the analysis is based on the assumption that the counts per channel have statistical uncertainties proportional to the square root of the counts. In fact the counts may be the difference between two large numbers and the uncertainty would be the square root of the sum of the numbers.

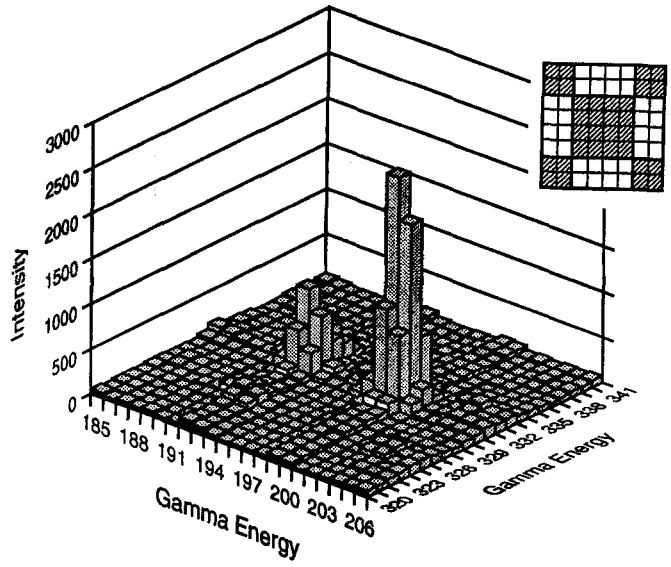

Fig. 1. Graphical representations of coincidence data and gating analysis. The left portion of the figure is a 22 channel by 22 channel section of a gamma-gamma coincidence matrix. Each channel is 1 keV wide. The number of counts in each of the 484 2D channels is given along the vertical axis. The right portion shows how gates might be combined to obtain the net counts in a peak.

Several advantages can be anticipated for multidimensional analysis. Excessive statistical noise is avoided. Direct analysis of the histogram prior to any subtractions permits uncertainties to be inferred from the data. Peak separations (diagonal) may be significantly larger in two dimensions. Finally, CHEMALY[7] and RADFORD[8] point out that the larger number of degrees of freedom in multidimensional spectra permits better tests of data models.

Even with large arrays of detectors, radioactive decay (fission excepted!) will yield primarily 2-fold coincidences, and these are the basis for most of the description of the method in the following sections. Some of the steps have been reported[9,10] and Radford's methods,[8] although only recently reported in detail, have been broadly used by in-beam spectroscopists and are known as "Radware".[11]

Peak search and fitting procedures

In many ways multidimensional fits are simple extensions of one-dimensional methods. There must be some definition of a peak, which must be sufficiently

different from non-peaks to permit detection by numerical methods. Regardless of the number of dimensions, we can separate analysis into peak search and peak quantification. The main difficulties with both peak detection and quantification of singles data are overlapping peaks and the lack of obvious background among closely spaced peaks. In higher dimensions both problems are less serious because peaks do not spread as rapidly as the background. There are more pure background regions and higher (peak)/ (local background) ratios.

One-dimensional fits

Major non-proprietary programs for analysis of 1D gamma-ray spectra include SAMPO,[4] GAMANAL,[5] and HYPERMET.[6] As preparation for our development of multidimensional analysis procedures, we have written a suite of programs[7] to analyze single parameter spectra. Since the methods are not new and the parts applicable to 2D analysis are given below, no details will be given here. In brief, the peak search technique is that developed by MARISCOTTI[12] and based on local extrema in the smoothed second difference. The peaks reported by the search program and those expected from the experimental context (e.g., the energy difference between two known levels) are taken over to a fitting program. Peaks that are close together ($\pm 5\ \sigma$) are fitted as a group. The group is described as a set of simple gaussians on a quadratic local continuum, and the best parameters are obtained by the Levenberg-Marquardt method.[13]

Two-dimensional analysis

In brief, the analysis consists of computing the continuum[9] and the ridges[10] in the full 2D spectrum, then searching for peaks in the difference spectrum (data minus continuum and ridges), and finally fitting each peak (or group of overlapping peaks) by a weighted least squares technique.

The peak search is based on numerical second differences calculated for positions i and j along the respective axes. Just as for one-dimensional data, the method can find isolated peaks and most multiplets, but cannot resolve the components of multiplets. This resolution is deferred to the fitting stage. After the peak search gives the preliminary coordinates for the coincidence peaks, a two-dimensional Levenberg-Marquardt method is used to refine energies and intensities. The function used to model the peaks is the product of two simple gaussians superimposed on the ridges and two-dimensional continuum found in separate steps of the analysis. In general the FWHM function for each axis is fixed during the fitting, but it can be fitted as well (such as to find effects which will broaden peaks

beyond the simple detector resolution. The fitted region includes $\pm 2\ \sigma$ along both i and j and is expanded to include any peaks within 4 σ in any direction. When the peak shape is not being sought in the fit, such as when the FWHM is assumed to be given by the function describing experimental resolution, the 'peaks' do not need to be assumed to be gaussian.

The location of a peak centroid is defined as the channel (i_0,j_0) where the smoothed second difference

$$ss_{i,j} = dd_{i,j} / sd_{i,j} \tag{1}$$

reaches a local maximum. The numerical second difference is calculated from

$$dd_{i,j} = \sum_{k=-l}^{l} C_k N_{i+k,j} + \sum_{m=-n}^{n} C_m N_{i,j+m} \tag{2}$$

and its standard deviation

$$sd_{i,j} = \left[\sum_{k=-l}^{l} C_k^2 N_{i+k,j} + \sum_{m=-n}^{n} C_m^2 N_{i,j+m} \right], \tag{3}$$

where N_{ij} is the number of counts at channel (i,j). The coefficients C_k an C_m are given by

$$C_k = \frac{-100\left(k^2 - \sigma_i^2\right)}{\sigma_i^2} \exp\left[-\frac{k^2}{2\sigma_i^2} \right] \tag{4}$$

and

$$C_m = \frac{-100\left(m^2 - \sigma_j^2\right)}{\sigma_j^2} \exp\left[-\frac{m^2}{2\sigma_j^2} \right] \tag{5}$$

with σ_i and σ_j are the appropriate gaussian parameters in the region. The series are terminated when the coefficients drop below $C_0/100$. With this termination the sum of the Cs is no longer zero, but the bias (0.3% of the sum of the absolute values) is small for data that rarely have counts with statistical errors of less than 1%.

The peak search gives preliminary coordinates for the peaks in the matrix. The function

$$peak_{i,j} = height \exp\left[-\frac{(i_0 - i)^2}{2\sigma_{i_0}^2}\right]\exp\left[-\frac{(j_0 - j)^2}{2\sigma_{j_0}^2}\right] \tag{6}$$

is used to model the peaks and is superimposed on ridges and continuum

$$y(i,j) = height \exp\left[-\frac{(i_0 - i)^2}{2\sigma_{i_0}^2}\right]\exp\left[-\frac{(j_0 - j)^2}{2\sigma_{j_0}^2}\right] + RI(i,j) + RJ(i,j) + C(i,j) \tag{7}$$

and compared with the data. The quality of the fit during application of the Levenberg-Marquardt technique is given by

$$x^2(\mathbf{a}) = \sum_{i=1}^{N}\sum_{j=1}^{N}\left[\frac{y_{i,j} - y((i,j);\mathbf{a})}{\sigma_{i,j}}\right]^2 \tag{8}$$

The adequacy of the model – in particular, the proper number of components and their energies and intensities – is inferred from the reduced chi square and a goodness-of-fit parameter which computes the probability that a chi-squared as poor as that obtained would occur by chance. Multiplets must be handled by adding more components to the model of the fitted region and repeating the optimizations.

Care must be given to testing the quality of the peak search results to avoid being trapped in a local minimum around a poor approximation.[7] In some cases one or more of the gaussians in the set to be optimized might be chosen because of possible transitions based on the best level scheme.

The functions which describe peaks need not be gaussians and can be different for the two axes. We have used the technique with coincidences between large Ge detectors (grouped to form on parameter) with small, high-resolution detectors (LEPSs). For x rays the function to be optimized was the peak shape for the four K components seen in moderate resolution (α_1, α_2, β_1, β_2). Discontinuities in channel size can be accommodated as well.

Higher dimensions

All of the techniques we have used for 2D spectra can be extended to three dimensions as long as the fitting region can be addressed during the computation. RADFORD[8] has described storage needs for "cubes" constructed from 3 and higher fold coincidence data. It is particularly important that when the analysis is driven by the goodness-of-fit between the predictions of a level scheme and the data, that both the data and the computed spectrum in the fitting region be in fast memory during the fit.

Interpreting data

Each two-dimensional peak yields a pair of energies and an intensity. Fitting a two-dimensional matrix yields a list of these peaks. This list is not easy to interpret. Even though enough components may have been added to give good fits throughout the matrix, some of the peaks may in fact be multiplets involving components with energies much further from the centroid than the statistical uncertainty in the centroid would suggest. It is hard to organize the list in such a way that associated peaks can be recognized. The list for three-fold coincidences would be even more complicated and difficult to interpret. RADFORD[8] has solved this problem by making the level scheme the primary reference and comparing it with data by computing gated spectra that experimentalists are accustomed to using. We also use the level scheme as reference (see below), but our comparison does not use gated spectra.

The power of the interaction data with models has been shown for HELMER[14] for single parameter data. His GAUSS VIII suite of programs combines nuclide libraries with conventional spectrum analysis routines to obtain state-of-the-art assays of radioactive materials.

Representation of models

As pointed out above, the data can be interpreted only in the context of some model. For radioactive decay the model is a combination of one or more decay schemes coupled to the detection system (energy and time resolution, detector efficiency, angular positions, inter-detector scattering, etc.). We have used the matrix representation of decay schemes[15] and have extended it to include level-to-level feeding patterns. As WAPSTRA[1] has shown, placement of a transition in a decay scheme can imply many coincidences in addition to those between transitions in and out of a given level. The intensities depend on branching ratios, conversion coefficients, and sometimes lifetimes. CHEMALY[7] has written X Windows routines

to display a matrix decay scheme and to allow selection and modification of particular transitions. The modified values are used to compute the coincidence spectrum, the spectrum is compared with the data, and the goodness-of-fit is compared with the goodness-of-fit before the modification. Since all of the coincidences implied by the placement are used in the comparison, incompatibilities are quickly recognized. RADFORD[8] has developed extensive interactive tools based on graphical representations of bands and interband transitions.

An important aspect of matrix representations of the decay schemes is that the upper limits of intensities of unobserved transitions are included in the schemes. It would be difficult to show these limits on a conventional lines-and-arrows scheme. Some of the limits have no value, such as the limit for a transition between states with very different spins, but those cases are easy to recognize (large upper limit on single particle strength) and do not expand the physical size of the scheme.

Discussion

Two rather different situations can arise in studies of radioactive decay by coincidence techniques. At one extreme the object might be to find low probability events in otherwise rather simple decay schemes. At the other extreme, the scheme might be so complicated that new levels or bands of levels await detection by improved techniques.

Our procedures[7,9-10] and those of RADFORD[8] were developed for complex coincidence data with many overlapping peaks in single-parameter projections. Analysis of spectra involves finding the relevant spectral components and determining their intensities. The process is not linear. Interpreting the results of analysis proceeds in stages, the most important being the construction of a fragment of gamma-ray cascades. Some peaks are obvious in the projections, others are predicted by existing level schemes, and others must be found by searching the N-dimensional space. Such searching requires a mathematical distinction between peaks and other features and benefits from treatment of the continuum before peaks are analyzed. Because the continuum is the sum of small contributions from a large number of different coincidences, it takes on rather simple average properties.

Different techniques would be appropriate for simple decay schemes. For example, the gamma-ray spectrum of ^{65}Zn is dominated by the 1115.6-keV transition from the second excited state of ^{65}Cu to its ground state. STELSON[16] measured the intensity of the 344.9–770.7 cascade (0.006% of the 1116-keV decays) by conventional coincidence techniques. If the measurement had been done by 2D spectrum analysis, the analysis could have concentrated on the 345–771 and

771–345 peaks. The continuum under the peaks (chance coincidences plus 1116-keV gamma rays scattered between detectors) could be determined by numerical interpolation from regions near the peak. In any event, the techniques used to compute the continuum in in-beam spectra would not apply here. Note that coincidence techniques could not give information on the intensities not associated with the cascades, such as decay following electron capture of ^{65}Zn directly to the 771-keV level. This limitation of coincidence measurements is analogous to the difference between relative and absolute gamma-ray intensities.

These techniques for multidimensional analysis are steps toward the "complete" spectroscopy HIRSKIND[11] anticipates for heavy ion reactions studied with large arrays of detectors (such as GAMMASPHERE[17] and EUROBALL[18]). An important aspect of completeness is the close interaction between models and data; the data cannot be interpreted without making many assumptions about the "physics", and the main flow of logic is to show that the data are consistent with a particular, albeit detailed, model. For high spin states, more complete spectroscopy will reveal non-yrast bands produced in low intensity and low intensity inter band transitions which are evident for intra band intensities but are individually below detection thresholds. For radioactive decay (again, fission excepted), 'complete' spectroscopy will involve relatively few additional levels but much more complete characterization of transitions among levels.

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