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Beta-decay total absorption measurements for nuclear technology and astrophysics

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Abstract. An accurate determination of the beta-decay intensity distribution is of importance for basic nuclear physics but also in the fields of reactor technology, astrophysics and fundamental interactions. Most of present information comes from experiments performed with high resolution low efficiency Ge γ-ray spectroscopy, which fails to locate the beta intensity at high excitation energies in complex decays. The total absorption spectroscopy technique with large 4π scintillation detectors can give the correct answer but requires an involved procedure in order to extract the information. Recent work on the systematization of the analysis methods and on the evaluation of the associated systematic uncertainties shows that reliable beta strength distributions can indeed be obtained.

1 Importance of beta-decay intensity and strength distribution measurements

An accurate determination of the distribution of the β-decay probability as a function of the energy of the excited level in the daughter nucleus is of importance in basic nuclear physics but also in other fields [1]. In basic nuclear physics the β-strength distribution can be utilised as a tool for nuclear structure studies since it is sensitive to the overlap of the initial and final wave functions and the basic interaction is well understood. The relation of the β-intensity I_β into a level of energy E_x with the β-strength S_β and the squared theoretical matrix elements B is given by:

$$S_\beta(E_x) = \frac{I_\beta(E_x)}{f(Q_\beta - E_x)T_{1/2}} = \frac{1}{D}B(E_x) \qquad (1)$$

where f is the Fermi rate function and D a constant. It should be noted that the Fermi function strongly enhances the role of the β-intensity at high excitation energies. From the comparison of the accurately measured and the calculated β-strength distributions one hopes to improve the theoretical models which, in turn, may have an impact on other areas. In nuclear astrophysics one key ingredient in the calculation of the neutron capture driven r-process nucleosynthesis are the β-decay half lives for nuclei along the r-process path. The path is largely determined by the competition between the neutron capture and the reverse process of photo-neutron emission, and their half lives have a strong influence on the final abundances of r-elements. Since the r-process occurs on the neutron-rich side and far away from the β-stability valley this information is mostly out of experimental reach and has to be determined from theoretical models of the nuclear structure and decay. The usual procedure [2] is to validate the theoretical extrapolations using the measured half lives close to stability. However as can be seen from equation (1) the half lives are inversely proportional to the integral over the full β-window of $f(Q_\beta - E_x)B(E_x)$. It is reasonable to expect that the predictive power of theoretical decay models far from stability would improve if they are able to reproduce the detailed β-strength distributions and not only the integrals. Thus β-strength measurements on neutron-rich nuclei far from stability, in particular systematic measurements along N or Z in selected regions should improve our knowledge of the synthesis of the chemical elements and the astrophysical site(s) of the r-process.

The β-intensity distribution itself may also have a practical significance. This is the case of the reactor decay heat. The heat liberated after the extinction of the fission processes in a reactor is due largely to the β-decay of the accumulated fission products. The evolution of the heat per unit time in the irradiated fuel can be obtained from a knowledge of the fission product inventory N_i (which depends on the reactor irradiation history), its temporal evolution (which can be computed from the decay laws) and the amount of energy liberated in the decay by the β-particles and the electromagnetic radiation (which depends on the β-intensity):

$$H(t) = \sum_{i=1}^{n} N_i(t)\frac{\ln 2}{T^i_{1/2}} \int_0^{Q^i_\beta} I^i_\beta(E_x)[E_x + \bar{E}_\beta(E_x)]dE_x \,. \qquad (2)$$

The above simplified relation assumes that for the decay to a specific level the γ-ray energy is equal to E_x and the β-particle energy $\bar{E}_\beta(E_x)$ is an average over the β-continuum. In fact such types of calculation are very useful since it is not always possible to make measurements of the decay heat evolution for a specific fuel composition and irradiation or simply because it is desired to study the behaviour of hypothetical ones. Therefore they are important in the context of reactor operation and safety assessment, waste management and facility design. Unfortunately there exists a long standing problem for this type of summation calculation based on evaluated decay data, namely a persistent discrepancy with benchmark measurements. There is strong evidence [3] that a major contributor to the discrepancy is the inaccurate knowledge of the β-intensity distribution used in equation (2). Improved measurements of these distributions will have a substantial impact on those fields.

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The bulk of the information on β-intensity distributions in the decay data bases comes from high resolution spectroscopy with Ge detectors which aims at the construction of decay level schemes from the detection of individual γ-rays. From the level scheme, the β-decay probability is extracted based on the γ-ray intensity balance. Since this technique relies on the detection of *all* γ-rays in each cascade, it is subject to error in the case of complex decays. For those decays involving a large number of levels, and due to the limited peak efficiency of Ge detectors, there is a tendency to miss the weak primary γ-ray transitions from levels at high excitation energies and therefore to assign excessive β-intensity to low energy excited levels. This systematic deviation is known as the *pandemonium* effect [4]. The severity of the problem depends on the level density accessible within the β-window, on the de-excitation pattern and on the β-intensity distribution itself. It has a greater effect on the decays with large Q_β-values and those where the β-strength concentrates close to the end. It is not possible to give a general rule, but our experience shows that one should regard as suspect the decay data for medium and heavy nuclei with Q_β more than $\sim$3 MeV.

The Total Absorption Gamma-ray Spectroscopy (TAGS) technique was introduced precisely to avoid this difficulty. It aims at the detection, with close to 100% certainty, of the γ-ray cascade de-exciting the levels populated in the daughter nucleus rather than the individual transitions. Such a high total detection efficiency can be obtained using large, close to 4π, scintillation detectors, since already for a cascade of two γ-rays the probability of both escaping becomes very small and it decreases with the cascade multiplicity. A high peak or full absorption efficiency is on the other hand more difficult to obtain and, contrary to the total efficiency, it decreases with multiplicity. Therefore in order to reconstruct the β-decay probability from the measured spectra the response of the spectrometer to the decay must be unfolded. This is not an easy task since it is necessary to know the response to all possible emitted radiation and combine them in the right proportion.

If one is interested only in the average γ-ray or β-particle energy released in the decay it is not necessary to rely on the correct β-intensity distribution, since it is possible to measure these quantities directly [5]. However a comparison of the average energies obtained in ref. [5] with those obtained by TAGS measurements in ref. [6] for up to 25 isotopes reveals large discrepancies [7]. On average, mean γ-ray energies in ref. [5] are 177 keV higher than in ref. [6], while mean β-particle energies in ref. [5] are on average 360 keV smaller than in ref. [6]. A careful analysis of both types of measurement shows that both might be affected by several sources of systematic deviation. In fact in ref. [5] they employ a consistency check of the results, namely that the sum of all three average energies (electrons, γ-rays and neutrinos) should add up to the Q_β-value, which shows deviations ranging from -600 keV to $+900$ keV for the isotopes under comparison. In the case of the average γ-ray energies the direct method of ref. [5] requires a normalization to well known decays, which was in their case ^{91}Rb. However the TAGS result in ref. [6] shows that it is affected by the *pandemonium* effect. If confirmed this will imply a renormalization of all average γ-ray energies of ref. [5]. This discussion reinforces the importance of the TAGS approach to average decay energies.

The complexity of the analysis of TAGS data is probably the reason why this technique has not become more popular. There is perhaps also a lack of confidence in the results as a consequence of the absence in the past of a detailed study of the assumptions required and the associated systematic errors. For this reason we have undertaken a systematic study of the issues related to the analysis of TAGS data. We describe in the next section recent results in this field together with the status of the technique and future developments.

2 Total absorption spectroscopy

2.1 The technique

The TAGS technique applied to β-decay measurements was introduced in a pioneering work at ISOLDE [8]. Their spectrometer consisted of two closely placed moderately large NaI(Tl) detectors with rather poor peak efficiency (28% at 1 MeV). A well-type spectrometer with 60% higher peak efficiecy was built at LNPI (St. Petersburg) [9] and later used at GSI (Darmstadt) and JYFL (Jyväskylä). An additional 45% increase in efficiency was achieved with the spectrometer installed at INEL (Idaho) [10]. A true 4π spectrometer with a peak efficiency of 65% at 1 MeV was built at LBL (Berkeley) and later transported to GSI (Darmstadt) [11]. Recently [12] a new spectrometer has been installed at ISOLDE with an efficiency slightly lower than the LBL device. All these spectrometers use one (or two) large crystals made of NaI(Tl) because it offers the highest energy resolution. The good resolution together with a high peak efficiency allows one to resolve not only the structure in the β-intensity distribution but also helps to locate and eliminate contaminations. On the other hand, the use of a multi-crystal spectrometer offers the opportunity to measure the γ-ray cascade multiplicity. This information might be useful during the analysis of TAGS data. A new spectrometer consisting of 12 crystals of BaF_2 (for good timing and low neutron sensitivity) is being commissioned by the Surrey-Valencia collaboration [13] and is planned for installation at ALTO [14]. Finally we want to remark that although a TAGS spectrometer will register both the γ-rays and the β-particle from the decay, it is not sensitive enough to the latter and the determination of the decay probability to the daughter ground state requires the use of an additional β-detector [15].

The most dangerous source of systematic error in the TAGS technique is the presence of background or contamination counts in the spectra. Since these detectors are large the count rate from the ambient background is also large, imposing a limit on the minimum activity which can be studied. The room background can be greatly reduced using appropriate shielding but it must still be subtracted from the measured singles spectra. Alternatively background-free spectra can be obtained by using ancillary detectors to tag the decay. These could be a thin Si or plastic scintillation detector which allows one to obtain the spectrum in coincidence with the beta particle. However the inherent electronic thresholds introduce an efficiency dependence close to the Q_β-value which is difficult to determine accurately. A combined analysis of both singles and coincidence data helps to overcome this difficulty.

This type of problem does not appear in the case of an X-ray detector which can be used to select the EC component for EC/β^+-decay. Additionally tagging with X-rays provides isotopic identification (see below). Inevitably the placement of ancillary detectors close to the source will adversely affect the detection efficiency. Electronic pulse pile-up represents a form of inherent background which is noticeable close to the end of the beta window and which must also be corrected for [16].

Isobaric contaminations are often unavoidable in the secondary beams from on-line mass separators where these spectrometers usually operate. Even for mono-isotopic beams the daughter activity must be considered. The conventional procedure to separate the different isotopes is based on their half lives. The use of a sample collection and transport system and the selection of appropriate collection and measuring times allows one to disentangle its composition. If the half lives of the different radioactive species are not different enough the separation becomes uncertain or simply impossible. In this situation chemical separation becomes mandatory for TAGS measurements since the lack of resolution does not allow any isotopic identification from the γ-ray energies. In some rare cases the separation can be provided by the chemico-physical processes in the mass separator ion source. An alternative is to force the selective ionization of a given isotope by coupling a laser with appropriate wave length to the ion source. The development of dedicated schemes of selection for specific elements has been pursued for example at ISOLDE (CERN), LISOL (Leuven), ISAC (Vancouver) and IGISOL (Jyväskylä) (see [17] and references therein). The isotopic purification of the beam could also be achieved by extreme mass resolution. Additional mass resolution at an on-line mass separator can be obtained by storing the secondary beams in ion traps operating in the appropriate mode. We have very recently performed an experiment at the University of Jyväskylä where for the first time a TAGS spectrometer was coupled to such a system. The JYFLTRAP [18] purification setup consists of a tandem of Penning traps after an RFQ ion cooler. This, together with the characteristic feature of the ion guide type separator IGISOL, allowed the measurement of several refractory element isotopes (Tc, Mo, Nb) of mass $A\sim100$ of importance for the calculation of reactor decay heat in the cooling time region $t\sim1000$ s. A particular advantage of this purification method is its universality, which is limited only by the attainable mass resolution.

In-flight separation of the reaction products from high energy reactions is an alternative method for the production of rare isotopes with very short half lives. In the future we are planning to perform TAGS measurements for selected isotopes of interest in nuclear structure and astrophysics at the international installation FAIR (Facility for Antiproton and Ion Research) which is planned at Darmstadt (Germany). The high energy ions coming from the Super Fragment Separator will be decelerated and directed to the DESPEC experiment [19] cave where they will be implanted in an active stopper. The stopper located in the middle of the spectrometer consists of a stack of position-sensitive Si detectors. The subsequent emission of a β-particle or X-ray will be registered in the same pixel in such a way that for each decay the separator will provide Z and A identification of the ion. The application of the TAGS technique in this case represents a particular challenge due to the large beam sizes (several cm) and the strong beam related background. Special emphasis will be put on measurements of neutron-rich nuclei where the process of delayed neutron emission becomes important when moving away from stability. This introduces not only the need to perform dedicated neutron measurements to complete the information on the β-strength distribution but it also means that care should be taken with the undesired contamination introduced by this process in the TAGS spectra. All of these considerations are being taken into account in the design of the spectrometer [13].

2.2 The analysis

In order to obtain accurate information about the distribution of the β-intensity in the decay from the measured spectrum we need to know accurately the response of the spectrometer to the decay into the excited levels of the daughter nucleus. In practical terms this requires a) the means to obtain the response to each of the emitted particles (β-particle, X-rays, γ-rays, conversion electrons...) and b) a knowledge of all possible cascade paths. The response R_{ij} to the electromagnetic cascade de-exciting level j can be obtained in a recursive way

$$R_{ij},\ R_{i-1\,j} = \frac{1}{2}\sum_{k=0}^{j-1}\sum_{l=0}^{i} b_{jk}\ g_{i-l\,j-k}\ R_{lk}, \qquad i = 0, i_{\max} \tag{3}$$

where b_{jk} represents the branching ratio for the transition $j \rightarrow k$ and $g_{i-l\,j-k}$ its response. At first it might seem that the second task is impossible to fulfill, since a complete knowledge of the decay level scheme can only be claimed for the lowest excited levels. But here one should remember that the principle of the total absorption spectroscopy technique is precisely to make the response weakly dependent on the cascade path (for an ideal spectrometer, i.e., one with 100% peak efficiency there will be no dependence at all). The response for the unknown part of the level scheme can be obtained from simple assumptions or by using the statistical model of nuclear decay. After obtaining the response R_{ij} to the decay one needs to solve by an appropriate method the inverse problem represented by:

$$d_i = \sum_{j=0}^{j_{\max}} R_{ij}\ f_j\,, \qquad i = 1, i_{\max} \tag{4}$$

in order to obtain from the data d_i the number of decays feeding each level f_j. Since the number of levels populated in the decay is in general very large some method has to be applied in order to reduce the dimensions of the problem. As one might suspect the analysis procedure is not straightforward. If we follow previous work we see that in the initial work at ISOLDE (CERN), and later at OSIRIS (Studsvik), the response functions to γ-rays were obtained empirically from source measurements using inter/extrapolations, the electromagnetic cascades were generated from the nuclear statistical model, and the solution of the inverse problem was carried out using the Gold-Scofield iterative method [20,21]. At LNPI (St. Petersburg) they obtained the β- and γ-ray response

functions using a Monte Carlo simulation code developed in-house, the user provided cascade branching ratios were introduced by hand and the deconvolution followed a simple "peel-off" method with an idealized response [9, 22]. Finally at INEL (Idaho) the individual responses were obtained with the code CYLTRAN, the level scheme for the construction of the decay response was introduced by hand and the decay feedings were obtained by a trial and error method until visual agreement with the spectra was obtained [23]. It is seen that in order to handle the analysis problem in those previous works several assumptions and simplifications were introduced but there is no discussion of the systematic uncertainties associated with them.

Given the situation we decided to undertake a systematic investigation of the different issues appearing in the analysis of TAGS spectra and the reliability of the extracted results, bringing the methods up-to-date. In this way we have shown [24] that modern Monte Carlo simulation codes for particle transport and interaction (such as GEANT3, Geant4...) using a detailed description of the geometrical setup and including the light production mechanism, are able to reproduce with great accuracy the spectrometer response to γ-rays and with slightly less accuracy the penetration of low energy β-particles. The latter fact suggests the convenience of introducing β-absorbers surrounding the source. We also studied [25] the suitability of different algorithms, well established in other fields, for the solution of the TAGS linear inverse problem represented by equation (4). As a result we concluded that the Maximum-Entropy and the Expectation-Maximization iterative algorithms are both well suited for the deconvolution of TAGS spectra, while the Linear Regularization algorithm shows a tendency to produce non-physical results in regions of low statistics. Moreover we could show that the results of the algorithms are stable upon variations of intrinsic parameters (number of iterations, regularization parameter...) and more importantly that the results of the different algorithms agree within a few percent in spite of their very different principles and concrete implementation. The most critical issue is perhaps the influence in the result of the lack of knowledge on the actual decay cascade path. In order to quantify this effect we have built a fictitious nucleus with a complex decay using the nuclear statistical model with known β-intensity and cascade branching ratios. In this way we can compare to the true β-intensity, the β-intensity extracted using different assumptions about the branching ratios for building the decay response. From this study we could conclude [26] that, as expected, the results are relatively insensitive even to rather unrealistic branching ratios. The β-intensity distribution is well determined (within a few percent) at high excitation energies while at lower energies, where the level density is low, there is a stronger dependence on the cascade assumptions. The total β-strength however is always determined within a few percent. The same procedure of varying the assumptions about the branching ratios should be applied to measured data in order to obtain an estimate of the systematic uncertainty. The use of experimental information about the γ-ray cascade multiplicity may serve to reduce the value of this uncertainty. During this investigation it also became clear that the procedure of grouping the response from several levels into one energy bin applied to equation (3) in order to reduce the dimension of the problem to a tractable size, introduces a sizable effect on the result. This effect is due to the fundamental impossibility of rebinning the branching ratios. We are currently investigating ways of circumventing this problem.

3 Conclusions

Total absorption spectroscopy is an essential tool for the determination of β-decay intensity distributions, particularly for nuclei far away from the valley of β-stability. The applications of this technique range from nuclear structure problems to astrophysical processes, nuclear technology and fundamental interactions. The accuracy of the results is an important pre-requisite for the usefulness of the technique. The development of new schemes of isotopic purification of the radioactive species at several installations will serve to reduce the risk of erroneous results due to contamination. The recent development of well established procedures for the analysis of TAGS data will allow the user to keep the systematic uncertainties under control.

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