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COLEGRAM, a flexible user-friendly software for processing of ionizing radiation spectra

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Alpha spectrometry, beta spectrometry, gamma-ray spectrometry, X-ray spectrometry

Abstract

COLEGRAM is a freely available spectrometry software dedicated to the study of ionizing radiation spectra. It can handle alpha, beta, gamma and X-ray spectra, proposing twenty-eight specific peak shapes in order to fit complex spectra using the non-linear least-squares fitting method. Different optimization cost functions can be used to perform the mathematical adjustment, which is guided by a user-friendly interface. COLEGRAM is also able to perform batch processing on a large number of spectra copying fitted peak information, based on a predefined model. General characteristics of the code and typical application examples are given to illustrate its potential.

1 Introduction

In the field of atomic or nuclear spectrometry, such as radionuclides characterization or X-ray fluorescence analysis, identification and quantification are generally based on energy-dispersive spectra, for which the result is represented as a histogram. In such cases, the histograms present structures like peaks, which can be routinely analyzed by commercial software. Nevertheless, more specific software is needed when the peak shapes are varied and require more detailed analysis.

Indeed, in alpha-, beta-, gamma- or X-ray measurements, the common detection method is based on measuring the energy of each detected event and counting these over a certain duration. The result of this measurement is a histogram, the spectrum, that presents different features representative of the emitted radiations and of the detector properties. The analysis of these spectra can be difficult as the evaluation of the different contributions must be distinguished individually and deconvoluted from the detector response function. Due to the physical process of interaction in the detector, the more general effect on a single-energy radiation is a Gaussian broadening of the corresponding emission line in the spectrum. Moreover, other physical effects, depending on the nature of the ionizing radiation have to be taken into account to answer the specific cases encountered in X-ray fluorescence analysis or alpha particle spectrometry, for example. There are several commercial software packages available for spectrum analysis (e.g., Maestro/GammaVision[®], InterWinner[®] or Genie2000[®] for gamma spectrometry, Visu-Alpha[®] for alpha spectrometry and PyMca [1] for X-ray analysis), which perform data processing most users may need. They are mainly based on automatic procedures and predefined libraries. However, more detailed spectrum analysis, without a priori knowledge of parameters of some components (i.e., energy, emission intensity or detector response function with secondary effects) may be required for specific studies.

COLEGRAM, a piece of software initially dedicated to gamma-ray spectrometry, was developed at the Laboratoire National Henri Becquerel (LNHB) to fulfill the need of a dedicated tool with a user-friendly interface. It was later extended to the analysis of other ionizing radiation spectra (alpha, beta, X) [2]. COLEGRAM offers to the user a wide choice of peak shapes in order to manage difficult to process spectra. The user can lock some fitting parameters or impose some constraints, which is generally not possible with commercial softwares. It has

1 been successfully used for many years at LNHB for specific metrological studies on radionuclide decay schemes. For
2 example, recently, Rodrigues and Loidl [3] used COLEGRAM to study high-resolution L X-ray spectra of ^{241}Am
3 obtained with a metallic magnetic calorimeter and revealed the presence of intense L X-ray satellites.

4 The new version of the COLEGRAM software is an improvement of the first release. In response to users'
5 feedback, it includes new peak functions, new graph options and new capabilities, such as batch processing to
6 automatically process a large number of similar spectra, to improve the functionalities and the field of use of the
7 software. Detailed information of the software are given in [4], here only the most important features of COLEGRAM
8 are mentioned and various examples are given to illustrate its potential.

9 2 General characteristics of COLEGRAM

10 The aim of the COLEGRAM software is to process experimental spectra coming from energy-dispersive detectors,
11 either semiconductor or other types (e.g., magnetic micro calorimeters, wavelength-dispersive spectrometers, etc.)
12 as long as the spectrum is a histogram presenting peaks (continuous beta spectra can also be processed). It is
13 written in C++ object-oriented language, making the executable software run quickly and efficiently. It runs under
14 any Microsoft Windows x64 environment and does not require any specific installation (only some dynamic link
15 libraries (dll) may need to be installed).

16 It is important to emphasize that the spectra processing is not automatic and requires a certain experience to
17 provide accurate results, which is facilitated by the user-friendly interface; in all cases, the user must be aware of the
18 relevant fitting parameters (peak position, tails, full-width at half maximum...) since no physical insight, neither
19 library, underpins the mathematical fitting procedure.

20 The spectra under study are typically composed of overlapping peaks superimposed on a continuous background,
21 where the information of interest is generally included in the peaks position (energy) and net area (quantification).
22 To derive these, the individual components are therefore modeled using mathematical functions, according to the
23 user's theoretical knowledge of the spectrum components. Fitting a mathematical function to n experimental data
24 points (x_i, y_i) consists of determining the optimum set of m parameters $(\vec{a} = a_0, a_1, \dots, a_m)$, such that the computed
25 values of the function $f(x_i, \vec{a})$ will be as close as possible to the experimental values y_i . The different parameters
26 of the function are obtained by the non-linear least-squares fitting method [5].

27 Generally, if Φ is the function to minimize, where σ_i represents the standard deviation of experimental value
28 y_i , three options are proposed for conducting the fitting. The first option corresponds to a simple minimization,
29 without taking into account any weighting of the experimental data ($\sigma_i = 1$): this is the "least squares" option:

$$\Phi = \sum_{i=0}^n [f(x_i, \vec{a}) - y_i]^2 \quad (1)$$

30 The second option uses the " χ^2 " criterion, i.e. including the standard deviation of each experimental point ($\sigma_i^2 = y_i$):

$$\Phi = \sum_{i=0}^n [(f(x_i, \vec{a}) - y_i) / \sigma_i]^2 \quad (2)$$

31 The last one uses the Poisson cost function which is adapted to low count spectra:

$$-ln(\Phi) = \sum_{i=0}^n [f(x_i, \vec{a}) - y_i \times ln(f(x_i, \vec{a}))] \quad (3)$$

32 The peak parameter values are obtained by minimization of Φ (or $-ln(\Phi)$), i.e., by canceling out the values of
33 the partial derivatives. This minimization is performed using the Levenberg-Marquardt algorithm which uses two
34 different approaches for the fitting procedures: far from the minimum, the gradient method is first used, then, closer
35 to the minimum, it is progressively replaced by an approximation of the Hessian method using only first derivatives.
36 In both cases, solving the equation system requires computation of the first derivative for each of the m parameters.
37 Peak areas are derived by integration of the mathematical function, using the fitted parameters.

3 Presentation of the user-friendly interface

The software working principle is based on the knowledge of the user who must configure the adjustment procedure. The main window proposes several menus and shortcut buttons (Figure 1). As a first step, the display of the whole spectrum allows the “Regions Of Interest” (ROI) to be determined, including selected peaks and some background (in red in the upper panel of Figure 1). The user has the choice to keep or remove the background of the ROI using the internal background evaluation method, as described in section 4.1.

Shortcuts buttons can open a spectrum file, define or erase a ROI, display it and change the scale in the active window. On both the complete spectrum and the ROIs, the experimental data can be displayed on a linear, square root or logarithmic scale. To plot the experimental data, points, circles, squares, triangles, lines and filled areas are available. Different colors can be chosen by the user for the background, experimental data, selected ROIs and calculated peak functions. Calibrating the spectrum for energy is also possible and can be performed in two ways: either by filling in channels versus energies or by filling the calibration curve parameters. Linear or second-order polynomial calibration can be used.

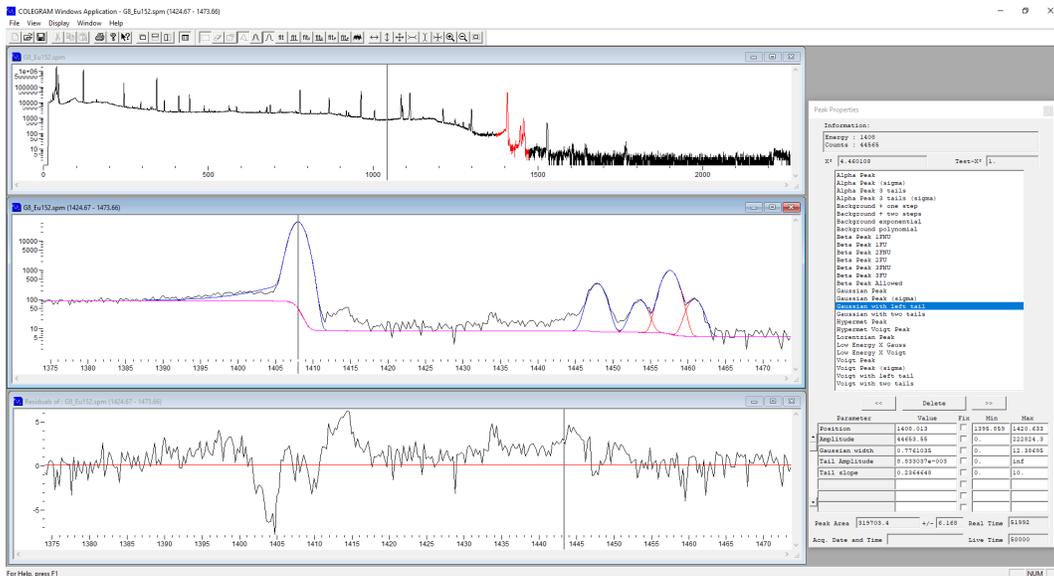


Figure 1: Colegram user-friendly interface. The left panel shows (from top to bottom) the whole spectrum, a selected ROI and the residuals of the fitting. The right “Peak Properties” floating window displays the peak parameters of all peaks present in the active ROI.

Once the user has selected a ROI in the initial spectrum to analyze, this can be displayed as the active window on the front, allowing the user to perform peak fitting (middle panel of Figure 1). A lot of room is given to the user as no physical model underpins any type of spectra. The “Peak Properties” floating window is open by default and displays the values of all the peak parameters and their associated constraints (minimum and maximum allowed values) for the active ROI window (if several ROIs are defined) (right panel of Figure 1).

The user can preselect a default peak shape type among the list of the types available and by clicking anywhere in the ROI window, a peak with the default shape is created at the position of the cursor. COLEGRAM calculates a first approximation of the amplitude and width of the new peak by making a small interpolation over the neighboring points. For example, the Gaussian width is approximated from the experimental channel contents around the cursor position, and the other parameters are set to default values. At any time during the fitting procedure, a parameter value can be adjusted or fixed, the peak shape, peak parameters and associated constraints can be changed from the “Peak Properties” window.

To adjust the mathematical function to the experimental data, the user can choose between three optimization cost-functions (least-squares, reduced χ^2 and Poisson), which should be sufficient for most common cases. All the

1 fitted parameters are accessible in the floating window and can be constrained individually, avoiding non physical
 2 results. To control the goodness of a fitting procedure, the χ^2 (Eq. 4) and an Adjusted- R^2 (Eq. 5) are calculated
 3 for each optimization, where n is the number of experimental data and m the number of fitting parameters).

$$\chi^2 = \frac{\sum_{i=0}^n [(f(x_i, \vec{a}) - y_i) / y_i]^2}{n - m - 1} \quad (4)$$

$$\text{Adjusted} - R^2 = \frac{\sum_{i=0}^n [(f(x_i, \vec{a}) - y_i)]^2}{\sum_{i=0}^n [f(x_i, \vec{a})]^2} \times \frac{n - 1}{n - m - 1} \quad (5)$$

4 The least-squares method provides the best result in most cases. In any fitting process, the χ^2 value is calculated
 5 and presented in the ‘‘Peak Properties’’ window together with the estimator of its quality (*Adjusted - R²*) which
 6 should converge to 1 when the fitted function is correctly representing the experimental data. COLEGRAM can also
 7 display the residuals in a separate window as shown in left lower panel of Figure 1. The corresponding peak areas
 8 are calculated according to the theoretical function using the fitted parameters and are shown in the same floating
 9 window. According to the Guide to the Expression of Uncertainty in Measurement [6], the uncertainty associated
 10 to the peak area is a combination of the standard uncertainties associated with each parameter. Uncertainties
 11 (variances) associated to each parameter are calculated during the fitting procedures, as well as covariances between
 12 the different parameters. The uncertainties associated with the peak areas are calculated according to the theoretical
 13 expression of the latter, i.e., the uncertainties associated with each fitting parameter are propagated according to
 14 the peak area expression to derive the combined uncertainty. As an example, the peak area of a Gaussian is
 15 $A \times \sigma \times \sqrt{2\pi}$ (A is the amplitude, σ the standard deviation), thus the associated absolute combined uncertainty is
 16 $\sqrt{(A \times u_\sigma)^2 + (\sigma \times u_A)^2} + A \times \sigma \times \text{cov}(A, \sigma) \times \sqrt{2\pi}$, where u_i is the absolute uncertainty of parameter i . In the
 17 present version, the calculated uncertainty associated with the peak area does not take into account the counting
 18 statistics, i.e. \sqrt{N} where N is the number of counts in the peak, to let the user appreciate the quality of the fitting.

19 The results can be saved in several formats, whether the user wants to keep the fitting parameters together
 20 with the spectrum or needs only the fitting results. COLEGRAM saves the entire spectrum with all the ROIs and
 21 the parameters of the fitted peaks in its dedicated ascii format (.spm) which can be read and processed again with
 22 COLEGRAM. It can also save only the fitting results as a spreadsheet (.csv), giving the choice to record only peaks
 23 positions and areas with their associated uncertainties or all peak parameters.

24 4 Fitting capabilities and examples of peak types

25 COLEGRAM can fit several types of spectra with basic functions such as Gaussian or Lorentzian. More specific
 26 shapes are dedicated to alpha or beta spectra. Some additional features are also available in order to correct for
 27 detection artifacts when a peak shape diverges from the ideal case. Each additional feature on an already existing
 28 peak shape is proposed as a new peak type. The available options are tails on both the low or high energy sides, step
 29 functions or shared Gaussian widths. The Voigt function, convolution of a Gaussian with a Lorentzian, is available
 30 and very useful for X-rays as the elemental (natural) linewidths become perceptible with modern spectrometers.
 31 The Hypermet function, introduced by Philips and Marlow [7], is a Gaussian summed with the convolution of a
 32 Gaussian with a left-tail and a step-function. Moreover, COLEGRAM proposes some derived forms of the Hypermet
 33 function: the Gaussian can be replaced by a Voigt or a double-step can be added as introduced by Scholze and Ulm
 34 [8].

35 In the case of alpha spectra, one left-tail is generally not enough to describe the energy-loss of the alpha particle
 36 in materials of different thicknesses or densities. Alpha peaks are best described by Gaussian functions that include
 37 two or three left tails [9, 10, 11].

38 Beta functions are also available in order to model the continuous spectrum component that comes from the
 39 detected beta particles. However, these functions represent the theoretically emitted spectrum from the nucleus,
 40 for allowed, 1st, 2nd, 3rd forbidden unique and non unique transitions only [12]. The user must be aware that the
 41 shape of the energy spectrum of the detected beta particles is highly dependent on the geometry, the materials and
 42 the non-linearity of the apparatus. Indeed, beta particles scatter very easily in matter and usually deposit only a
 43 small fraction of their kinetic energy in the detection system. The detected spectrum thus tends to be strongly

1 distorted compared to the initially emitted spectrum, with a significant increase of beta particles detected at low
 2 energy and a corresponding decrease of beta particles detected at high energy. In order to extract experimental
 3 form factors with COLEGRAM, the user must be aware that it is necessary to correct the acquired spectrum from
 4 the apparatus prior to any fitting.

5 Some background functions are also available but are only mathematical expressions (e.g., polynomial or ex-
 6 ponential) as no physical meaning supports their parameters. In summary, twenty-eight different shapes can be
 7 selected by the user to meet the needs of the regarded applications.

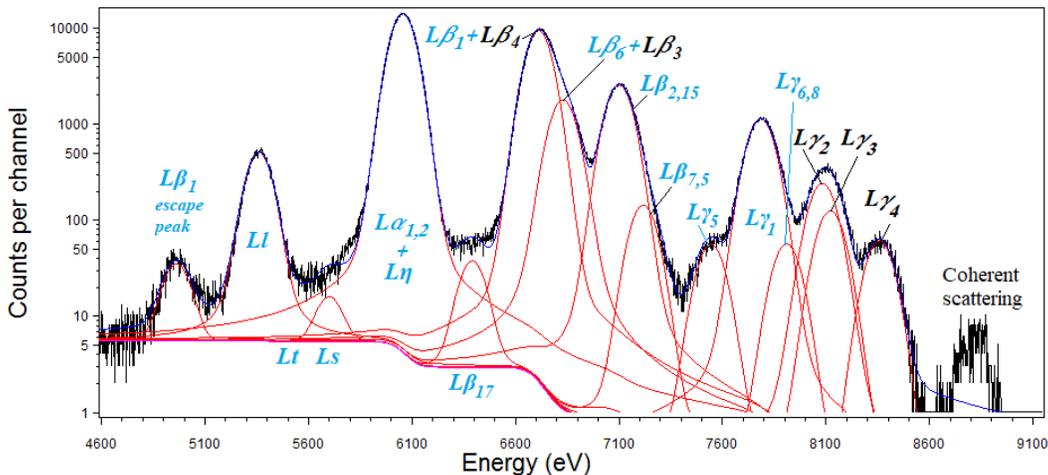


Figure 2: Gadolinium L lines for excitation energy E_0 with $E_{L_1} \leq E_0$. The experimental spectrum is shown in black, individual peak fits are shown in red, and the combined spectrum (sum of the individual fits) is shown in blue. All lines are fitted with “Voigt(Sigma)” functions which include a shared energy-dependence on the Gaussian width.

8 4.1 Discarding the background contribution

9 In a typical X- or gamma-ray spectrum, the peaks have variable amplitudes and a significant background is also
 10 present. We consider that the important information for the user is contained in the peak intensities and that the
 11 background contribution can generally be discarded. Several models can be used to compute the background under
 12 peaks: the simplest method fits a linear function to the limiting channels of the region of interest (ROI). A more
 13 sophisticated model would compute a polynomial, but these methods lack physical meaning. In COLEGRAM, we
 14 take into account the physical peak presence by an integration over the ROI channels [13]: this processing allows the
 15 step corresponding to the maximum energy of the full-energy peak to be included. If the user chooses to estimate
 16 the background with the internal COLEGRAM function [13, 2], it appears as a pink line in the ROI window (Figure
 17 1) and its contribution is subtracted from the ROI data values for the fitting procedure. Nevertheless, if the user
 18 needs a different strategy, some background functions are also available and can be fitted simultaneously with the
 19 peak functions.

20 4.2 X-ray spectra

21 In recent work, we used photo-ionization to study the L lines of gadolinium recorded with a silicon drift detector
 22 (SDD) in order to determine the partial fluorescence yields [14]. If some lines were too close to be distinguished,
 23 the hidden lines can still be taken into account provided that their amplitude and position are fixed. In this case,
 24 it means that their amplitude has to be evaluated independently. For example, at excitation energies larger than
 25 the L_1 absorption edge, all the L lines are visible (Figure 2). The lines were fitted with Voigt functions in order to
 26 take into account the Lorentzian width of the atomic levels responsible for the transition. All the Voigt peaks used

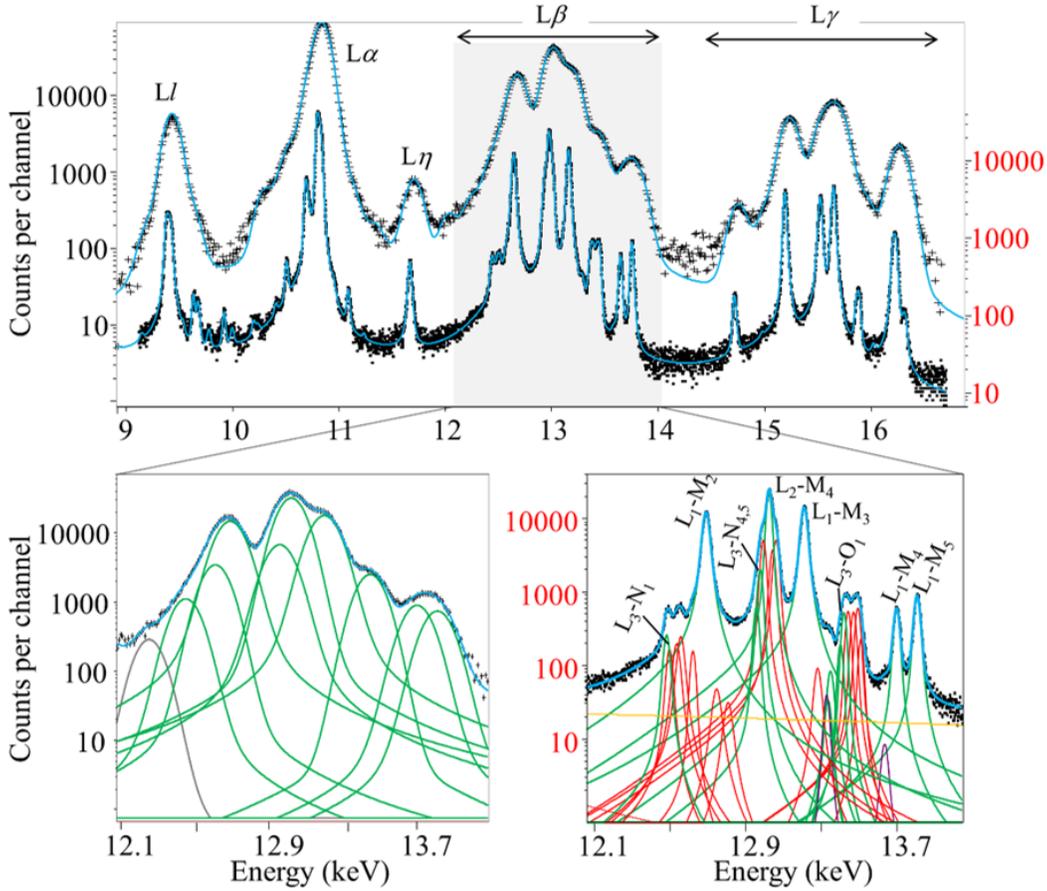


Figure 3: ^{210}Pb L lines. Upper figure: spectra obtained with an HPGe detector (cross marks and black y-scale) and with a cryogenic detector (square marks and red y-scale). The two lower spectra are a comparison of the L X-ray processing of the HPGe spectrum (left) and the spectrum acquired with the cryogenic one (right) in the $L\beta$ region. The green Voigtians are the diagram lines, the red Voigtians are the satellites, the purple Voigtians are L X-rays from Po, the gray Voigtian is the L X-ray from Pb (lead shield) and the orange line is the background. The blue lines are the resulting fits (courtesy from [3]).

- 1 have common parameters of Gaussian width, i.e., the energy dependence of the standard deviation follows a linear
- 2 law with photon energy, a feature which is also available for other peak shapes.
- 3 In the case of a radionuclide decay, the gamma rays and X-rays are emitted together and depending on the
- 4 detector resolving power, different lines can be distinguished or not. In Figure 3, we illustrate the study of the
- 5 X-rays emitted consecutively to the decay of ^{210}Pb , acquired with two types of detector: a conventional high-purity
- 6 germanium (HPGe) spectrometer and a cryogenic detector, characterized by a very high resolution allowing it to
- 7 distinguish between atomic characteristic lines (single ionization) and potential satellites (multi-ionization) [3].

8 4.3 Alpha particle spectrum

- 9 As another example, we focussed on the spectrum of ^{226}Ra and daughters measured with Si-PIPS detector (Figure
- 10 4). ^{226}Ra and its decay products emit alpha particles in cascade that are due to the decay of ^{226}Ra , ^{222}Rn , ^{218}Po ,
- 11 ^{214}Po and ^{210}Po . Alpha peaks appear as Gaussian shapes with a strong left-tail due to the high probability of
- 12 energy-losses of the particles before reaching the active zone of the detector. The left-tail is modeled with three
- 13 exponential functions having decreasing slopes.

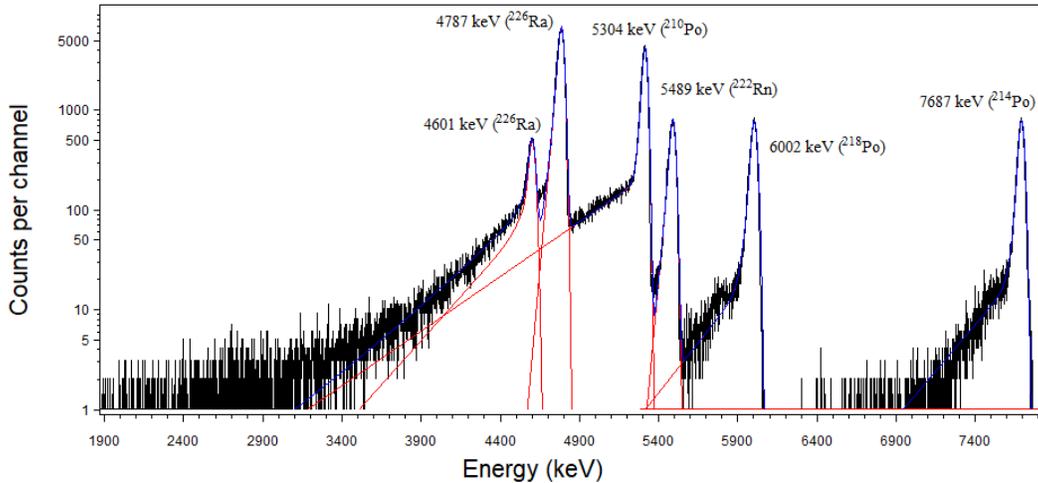


Figure 4: ^{226}Ra spectrum analysis. The experimental spectrum is shown in black, individual peak fits are shown in red, and the combined spectrum (sum of the individual fits) is shown in blue. All lines are fitted with “Alpha 3 tails”.

1 4.4 Batch processing

2 In some cases, the user may wish to process similar spectra with identical peak functions. In this case, a represen-
 3 tative spectrum must be processed and saved first in order to serve as a model for the other spectra. COLEGRAM
 4 uses all the defined peaks and associated free parameters or constraints (relaxing constraints on the amplitude
 5 though) as input for the other spectra. When saving the results as a table file using the same sub-menu, the user
 6 can choose how much information is to be saved: all information about the peaks or only the most relevant ones
 7 (i.e., the peak type, position and area). An example of the automatic processing of the XK lines of ^{103m}Rh in order
 8 to determine the radionuclide half-life is given in Figure 5. All the spectra were analysed using a two Gaussian
 9 peak shape as the model for the $K\alpha$ and $K\beta$ lines of rhodium and COLEGRAM automatically performed the same
 10 fitting to all of the spectra. Here, the decay time was found to be 56.15(6) min which is in very good agreement
 11 with the value in the literature (56.115 (6) min [15]), when considering the use of photon spectrometry for this type
 12 of measurement as compared to other more precise methods.

13 5 Conclusion

14 We presented the COLEGRAM software developed at LNHB, which can be useful in many application fields as
 15 long as the experimental result is a histogram showing peaks that contain useful information. At LNHB, we have
 16 been using this software for many years in the field of ionizing radiation to study the emissions of radionuclides
 17 including gamma- and X-rays. The accuracy of the fitting methods are of great importance when considering these
 18 spectra from a metrological point of view. The numerous available peak functions can meet most common needs in
 19 ionizing radiation. As a recent development, batch processing now allows a large number of similar spectra to be
 20 processed, which is particularly useful when the information contained in each spectrum is varying based on some
 21 external parameter (time, movement,...). Finally, COLEGRAM is freely available upon request from the LNHB.

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 24 fruitful discussion and help.

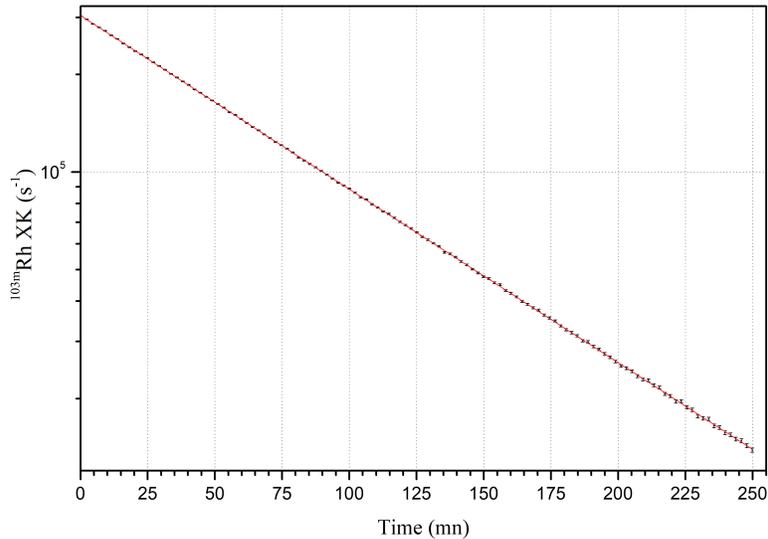


Figure 5: ^{103m}Rh spectra analysis: the batch procedure automatically fitted the XK lines of 120 spectra with the peak shapes and parameters from a model to calculate the corresponding peak areas.

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