Measurement of $\beta^-$-decay continuum spectrum of $^{138}\text{La}$

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Abstract – The LaBr$_3$:Ce scintillator offers the unique opportunity to study the $\beta^-$ radioactive decay of $^{138}\text{La}$. The $^{138}\text{La}$ isotope is one of the rarest isotopes on Earth (it is present as 0.09% in natural lanthanum) and because of its extremely long lifetime, of the order of $10^{11}$ years, large amounts of $^{138}\text{La}$ are needed for the measurement of the $\beta^-$-decay spectrum. In the literature, only one dedicated experimental measurement is present and the results seem not to be reproduced by the nuclear theory. A second measurement of the $\beta^-$-continuum spectrum is presented in this work. For this measurement, two LaBr$_3$:Ce scintillators ($3'' \times 3''$) and two different experimental techniques were used. The shape of the $\beta^-$-continuum spectrum, measured down to the energy of 10 keV, is found to be very similar to the one previously measured and published, but it is different from the theoretical published spectrum.

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Introduction. – This work is focused on the measurement of the energy spectrum of the electron emitted in the $\beta^-$ decay of the $^{138}\text{La}$. This is the second dedicated measurement ever done so far. The first measurement was performed in Delft by Quarati et al. [1]. The results of the first measurement, obtained with two different experimental techniques, are not in agreement with the theoretical prediction. This work aims to provide a second measurement and therefore to better understand the difference between the theoretical and the experimental spectrum. Similar deviations were also observed in other isotopes in earlier experiments, e.g., by Wu [2] and, they can be noticed more recently, in the spectra of ref. [3].

In this work the $^{138}\text{La}$ radioactive decay is described in the next section. We discuss the experimental method used to extract $\beta^-$-decay continuum spectrum of $^{138}\text{La}$ in the third section. The used experimental set-up and the results of the data analysis are described in the fourth and fifth sections, respectively. The conclusions of the work are discussed in the last section.

$^{138}\text{La}$ radioactive decay. – Lanthanum is present in Nature with two different isotopes: $^{139}\text{La}$, that is stable, and $^{138}\text{La}$, that has a lifetime of $1.05 \cdot 10^{11}$ years. The isotopic abundance of $^{138}\text{La}$ is 0.09% of the natural lanthanum.

The $^{138}\text{La}$ isotope is an odd-odd p-nuclide produced by re-processing pre-existing seed nuclei created in the $s$- and $r$-processes. As a natural result, the $p$-process nuclei are neutron-deficient and extremely rare in natural abundance.

The $^{138}\text{La}$ decays by electron capture ($\epsilon$) into an excited state of $^{138}\text{Ba}$ with 66.4% probability and the remaining 33.6% by $\beta^-$-decay into an excited state of $^{138}\text{Ce}$ [4,5]. In both cases, $^{138}\text{La}$ decays into an excited state of the daughter nucleus with the consequent emission of one $\gamma$-ray, as shown in fig. 1. In particular, in the electron capture decay, one $\gamma$-ray of 1436 keV is emitted while in the $\beta^-$-decay process, one $\gamma$-ray of 789 keV is emitted. Both the $^{138}\text{La}$ $\beta^-$ radioactive decay and the $^{138}\text{La}$ electron capture are 2nd-order unique forbidden transitions, in both cases the decay starts from a $5^+$ state ending to a $2^+$ state. The $Q$-value of the $\beta^-$-decay is 1044 keV. The sum of the $\beta$-decay electron and the antineutrino energy is 255 keV ($E_{\text{max}} + \bar{\nu} = 1044$ keV $-$ 789 keV $=$ 255 keV).

After the electron capture, the electrons have to be re-adjusted, therefore there is an additional X-ray and Auger electron emission (shell refilling). In particular, there is an Auger electron emission of 5.6 keV if the captured electron

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Fig. 1: Left panel: the decay scheme of $^{138}$La (see text). Right panel: the electron capture shell refilling (see text).

is in the $L$ shells, while, if the captured electron is in the $K$ shells, there is a X-ray emission of 31.84 keV and an Auger electron of 5.6 keV.

**Experimental method.** Since only few years, large volume LaBr$_3$:Ce are available as scintillator detectors. These scintillators are characterized by an intrinsic activity. This activity has two origins: the contamination due to the radioactive isotope $^{227}$Ac and its daughters and the presence of radioactive isotope $^{138}$La [6–8]. The contamination due to $^{227}$Ac and daughters manifests itself as an event with $\gamma$-ray equivalent energy above 1.5 MeV and below 3 MeV [6–8]. The $^{138}$La activity dominates the spectral region below 1.5 MeV. These large-volume scintillators contain an amount of $^{138}$La that is equal to its isotopic abundance. The used crystals were $3'' \times 3''$ and they contain about $10^{21}$ atoms of $^{138}$La, that is approximately 1.5 g. For this reason, LaBr$_3$:Ce detectors offer a unique opportunity to measure the $\beta^-$ continuum spectrum of $^{138}$La. Furthermore, the LaBr$_3$:Ce scintillator presents excellent scintillation properties. It has an extremely high light yield (63 photons/keV), the best energy resolution among scintillators (2.7% at 662 keV for small-volume crystals), excellent timing properties (300 ps of time resolution in small crystals) and a high density (5.1 g/cm$^3$) [6–25]. For this reason the LaBr$_3$:Ce scintillator can be used, both as the $^{138}$La source and as the detector to measure the $\beta^-$ continuum spectrum.

The measurement of the $\beta^-$ decay continuum spectrum of $^{138}$La, requires a coincidence measurement. In the $^{138}$La $\beta^-$-decay a $\beta$-decay electron, a $\gamma$-ray, and an anti-neutrino are emitted. Therefore, to properly tag a $\beta^-$-decay, a 789 keV $\gamma$-ray should be detected in one of the LaBr$_3$:Ce scintillators in coincidence with an event in the second scintillator, that is the electron emitted in the $\beta^-$-decay, as shown in fig. 2. The $^{138}$La electron capture decay constitutes a source of background. In fact, the 1436 keV $\gamma$-ray can be totally or partially detected in one of the LaBr$_3$:Ce scintillators in coincidence with the X-ray or the Auger electron in the second scintillator, as shown in fig. 2.

**Experimental set-up.** The $\beta^-$ continuum spectrum of the $^{138}$La was measured using two $3'' \times 3''$ LaBr$_3$:Ce detectors in coincidence, as shown in fig. 3. The detectors were coupled to two HAMAMASTU R6233-100sel PMTs with a standard HAMAMATSU voltage divider (E1198-26 and E1198-27). Two data sets were acquired, the first uses both analog and digital electronics while the second only digital electronics.

In the first measurement the coincidence was produced using a constant fraction discriminator (Ortec Quad CFD 935) and the time spectra was produced using a time to amplitude converter (TENNELEC TC 863). In this case it was possible to measure the spectrum starting from 30 keV, because of the CFD threshold. The two anode and the TAC signals were digitized using a 12-bit, 600 MHz LeCroy Waverunner HRO 66Zi oscilloscope. The acquisition trigger (trigger of the oscilloscope) was the TAC signal.

In the second measurement only the 12-bit 600 MHz oscilloscope was used. The coincidence was produced using the trigger logic of the oscilloscope, which requires that when one event is present in channel 1 of the oscilloscope a second event should be present within 150 ns in channel 2. In this case it was possible to measure the spectrum starting from 2–3 keV.
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The first measurement will be called “data set 1”, while the second one will be called “data set 2”. The acquisition time of the first data set was about 4 days, while the acquisition time of the second data set was about 8 days. The count rate was below 100 Hz.

**Results.** – The $\beta^-$-decay continuum spectrum of $^{138}$La was obtained from the coincidence matrix of the two detectors. The matrix, on the $x$-axis there is the energy measured in one of the detectors, while on the $y$-axis the energy measured in the other one is displayed. To reduce the random coincidences, the matrix was produced with a time condition of 4 ns around the prompt time peak, shown in fig. 4. The measured time resolution is about 1.1 ns. This spectrum is obtained from the detector anode pulses with a digital CFD algorithm. It is important to point out that the events outside the time condition of 4 ns around the prompt peak are less than 0.1% of the total.

The energy spectra of the two detectors were calibrated using standard $\gamma$-ray sources ($^{60}$Co and $^{22}$Na). The 789 keV peak was used to check the calibration and the possible drifts during all the measurement time. An important issue that must be taken into account is the scintillation non-proportional response, at low energies. It provides a visible distortion from a linear energy calibration of the spectrum. This non-proportional response was corrected by using the curve reported in [26].

The spectrum measured in one LaBr$_3$:Ce in coincidence with the events which deposited $\approx$ 789 keV is shown with a black line in fig. 6. It corresponds to the events in the white box of fig. 5. In the spectrum a peak at 37.44 keV (it is the sum between 31.84 keV that is the energy of the X-ray and 5.6 keV that is the energy of the Auger electron) and at 5.6 keV are clearly visible. They are due to the events associated with the Compton interaction of the 1436 keV $\gamma$-rays that are present below the 789 keV peak. As these transitions constitute the background their contribution has to be subtracted. The background spectrum (red line) is obtained using the events present in the red box of fig. 5 which, as those present below the peak at 789 keV, are produced by the Compton scattering of 1436 keV $\gamma$-rays. The electron spectrum, the blue line in fig. 6, is the difference between the black and red spectra.

As a cross-check, the $\beta^-$ spectrum of the two LaBr$_3$:Ce detectors for both data sets were compared (see fig. 7). The $\beta^-$-decay spectrum has the same shape in both detectors and for both data sets. The only evident difference is in the energy threshold: in data set 1 it was possible to measure only for $E > 30$ keV, while in data set 2 it was possible to measure for $E > 2$ keV. Therefore, for data set 2, the spectra of the two detectors were summed to increase the statistics as shown in fig. 8. From the data it is also possible to extract an estimate of the $L/K$ ratio. The $L/K$ ratio can be determined by comparing the area of the two Gaussians which fit the $^{138}$Ba de-excitation peaks (the 37.44 keV peak and the 5.6 keV peak).
We evaluated a ratio of 0.42 ± 0.03; this value actually relates to the \((L - M)/K\) ratio because the lower-energy Gaussian includes the events from the \(M\)-shell hole cascade in coincidence with the 1436 keV \(\gamma\)-ray. The value of the \(L/K\) ratio measured in [1] is 0.44 ± 0.04, that is in agreement with this value.

**Conclusion and perspectives.** – The measured \(\beta^-\) continuum spectrum, shown in fig. 8 by black line, is compared to the experimental data present in the literature (orange and red lines). The spectrum measured in this work has the same trend of the experimental spectrum reported in [1] obtained by the deconvolution method (red line) whereas the experimental spectrum obtained with the convolution method is slightly different from the one measured in this work. Both the measured electron spectra, however, have a very different behaviour with respect to the theoretical spectrum reported in [1].

The results of this work confirms the measurements performed in Delft and the discrepancy between the experimental data and the theoretical prediction of the \(\beta^-\) energy spectrum in the case of a second-order forbidden transition. From an experimental point of view, a new measurement will be performed exploiting the coincidence between a HPGe and a LaBr\(_3\):Ce detector. It will be possible to reduce the background below the 789 keV transition and therefore the incidence of time-correlated background. From the theoretical point of view, more detailed and accurate calculations are needed to verify if the shown discrepancy with the data is due to an incomplete understanding of the \(\beta^-\) decay process (in the case of second-order forbidden transitions) or to higher-order effects not included in the calculation of [1].

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