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Investigation of Electron Capture in ^{176}Lu with a LYSO crystal scintillator

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Abstract. The nuclide ^{176}Lu is one of the few naturally occurring isotopes that are potentially unstable with respect to electron capture (EC). Although experimental evidence for ^{176}Lu EC decay is still missing, this isotope is instead well known to β^- decay into ^{176}Hf with an half-life of about 38 Gyr. Previous searches for the ^{176}Lu EC decay were performed by using a passive Lutetium source coupled with an HP-Ge spectrometer. Our approach uses a LYSO crystal both as Lutetium source and as an active detector and the obtained ^{176}Lu EC branching ratio limits are 7×10^{-4} and 4.4×10^{-4} for the capture of the 1s and 2s electron, respectively. Previously known limits for the EC in ^{176}Lu are improved by a factor 3 to 15 depending on the considered EC channel.

1 Introduction

The naturally occurring isotope ^{176}Lu (2.6% abundance) it is known to β^- decays to ^{176}Hf with a half-life of ∼38 Gyr. However, ^{176}Lu is also one of the six naturally occurring isotopes that are potentially unstable with respect to electron capture (EC). In particular, evidence for EC decay was found for ^{40}K [1], ^{50}V [2, 3] and ^{138}La [1], but is still missing for ^{123}Te [4], ^{176}Lu [5] and ^{180m}Ta [6]. Figure 1 shows the decay scheme of ^{176}Lu: on the right side the dominant decay chain to ^{176}Hf is depicted, while on the left side there is the expected EC decay process to ^{176}Yb.

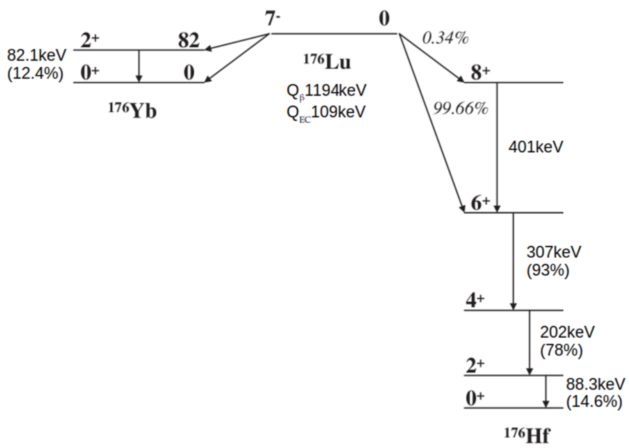

Figure 1. Decay scheme of ^{176}Lu [7, 8]. The reaction Q-value for EC decay of ^{176}Lu to the ^{176}Yb ground state is ≈ 109 keV. The percentages in the brackets are the γ-ray emission yields.

Previous searches for the ^{176}Lu EC decay were performed by using a passive Lutetium source coupled to a HP-Ge detector searching for the ^{176}Yb* 82 keV γ-ray or the characteristic Yb x-rays [5]. Our "active source" approach uses a 7.9g, 2mm thick, Lutetium–Yttrium OxyorthoSilicate (LYSO [9]) scintillator coupled to a R5946 PMT both as a ^{176}Lu source and as a detector placed on top of a GC2020 [10] HP-Ge detector. A passive shield made of 2.5mm of Cu and 5cm of Pb surrounds both detectors to minimize the environmental γ background. The measured energy resolution of HP-Ge detector was 1.65keV FWHM at 75keV and the measured energy resolution of the LYSO crystal was 20keV FWHM at 60keV. The energy scale of the LYSO crystal was calibrated with external ^{241}Am and ^{136}Cs sources, the known effect of the non-linearity of the LYSO Light Yield was taken into account in the evaluation of the measured LYSO energy. Further details on the experimental set-up are summarized in [11].

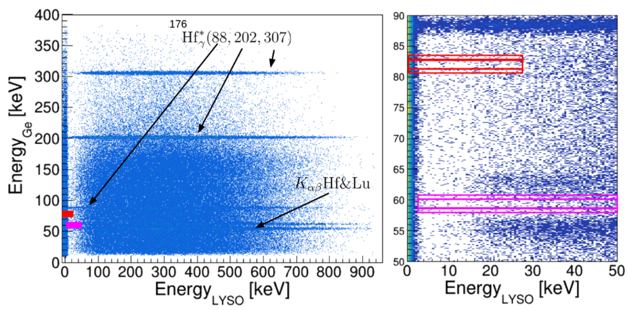

Figure 2. Energy$_{Ge}$ vs Energy$_{LYSO}$ spectrum and a zoom in the region of interest for EC. Red and magenta boxes are the regions of interest for ^{176}Lu EC to ^{176}Yb^{2+} and to ^{176}Yb^{0+} respectively. The zoom shows the $\pm1\sigma$ "Bulk" and $\pm(1\sigma$ to $2\sigma)$ "Sideband" regions considered in the analysis.

A total number of 1.5 Mevents with Energy$_{Ge}$ >15keV were acquired during 90h exposure time, most of the events collected by the HP-Ge detector come from the ^{176}Lu β-decay and the intrinsic/environmental back-

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ground. These two classes of events are well recognized in the bidimensional spectrum of FIG. 2. In particular, the population of events characterized by $\text{Energy}_{\text{LYSO}}$ <3keV (vertical strip in FIG. 2) are mostly due to intrinsic or environmental background. On the other hand, events characterized by $\text{Energy}_{\text{LYSO}}$ >3keV are mostly due to the continuous electron distribution emitted by ^{176}Lu β-decay. For this last class of events, in FIG. 2 are also visible the ^{176}Hf* γ-lines characterized by a fixed energy detected in the HP-Ge and the relative continuous distributions due to the Compton scattering. Similarly, at lower energy, the Hf x-rays due to internal conversion of ^{176}Hf* levels and the Lu x-rays due to photoelectric absorption, within the crystal, of ^{176}Hf* γ-rays, are visible. The rejection capability of the "active source" technique can be noticed in FIG. 2 where the red and magenta boxes are the regions of interest for ^{176}Lu EC to ^{176}Yb$^{2^+}$ and to ^{176}Yb$^{0^+}$ respectively.

In Figure 3 the energy distribution of the events measured by the HP-Ge is shown (magenta line). These data are plotted without any requirement on the LYSO measured energy, and as a consequence this plot is dominated by the background from the ^{176}Lu β-decay. This corresponds to typical landscape of the passive source measurement approach used in [5]. Profiting of the "active source" technique, most of ^{176}Lu β-decay events are rejected by the request E_{LYSO} <27keV (blue points).

This approach provides a factor ~20 in background suppression, and the resulting measured spectrum mostly corresponds to the intrinsic/environmental background characteristic of our HP-Ge detector. The black points on FIG. 3, are the dedicated measurement of the background events collected during 90h exposure time with the same set-up once the LYSO crystal has been removed. The x-rays of Pb and the characteristics γ-lines of ^{234}Pa* are identified in the background spectrum[1].

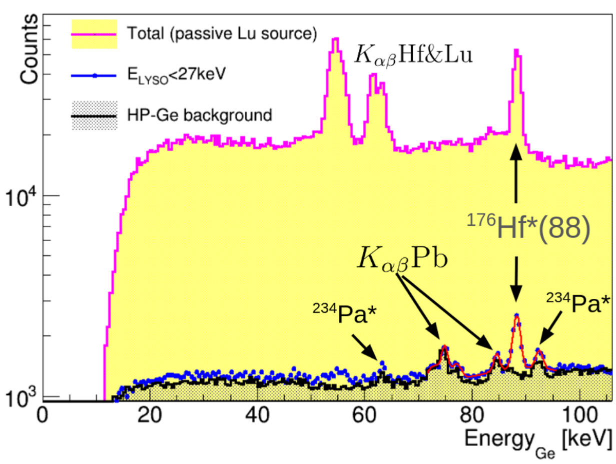

Figure 3. Energy distribution measured by the HP-Ge with no selections (magenta line) and selecting E_{LYSO} <27keV (blue points). A strong suppression of β decay background is achieved and the residual spectrum is dominated by the intrinsic/environmental background characteristic of our HP-Ge detector (black points).

[1]Natural radioactive chain of ^{238}U contains the ^{234}Pa isotope [12].

2 Search for ^{176}Lu EC/REC

The Q-value for EC decay of ^{176}Lu ($J^\pi = 7^-$) to the ^{176}Yb ground state ($J^\pi = 0^+$) is $\simeq$ 109 keV [8, 13, 14], and the one to the ^{176}Yb first excited state ($J^\pi = 2^+$) is $\simeq$ 27 keV. These EC decay branches, however, would be forbidden transitions of 7^{th} and 5^{th} order respectively. The relative branching ratios are then expected to be very small, also considering that forbidden transitions of order $>4^{th}$ have never been experimentally observed so far [15]. In these cases of highly forbidden transitions, that are suppressed by a large variation of the nucleus spin as compared with the sum of the angular momentum of the captured electron and of the emitted neutrino, an important channel for the decay is provided by the Radiative Electron Capture (REC) [16]. In this process an x-ray can be radiated either by the captured electron (Internal Bremsstrahlung) or by the father/daughter nucleus (Detour Transitions). This photon contributes with his spin to the overall angular momentum conservation. The REC decay has been experimentally observed for the forbidden decay of: ^{41}Ca, ^{59}Ni, ^{81}Kr,^{137}La and ^{204}Tl [17–21].

2.1 Search for ^{176}Lu EC to ^{176}Yb$^{2^+}$

Considering the 5^{th} forbidden EC transition of ^{176}Lu to ^{176}Yb$^{2^+}$, a strong identification signature is the 82.1keV de-excitation γ-ray observed in the HP-Ge detector. The maximum neutrino energy for this decay is $E_{82}^{\nu_{max}} = Q_{EC} - E_b - 82.1\text{keV}$, where E_b is the Yb atomic binding energy of the captured electron [22]. $E_{82}^{\nu_{max}} \geq 0$ therefore the capture of the 1s electron is not energetically allowed for this detection channel. The energy emitted in the Yb atomic shell rearrangement along with a possible REC x-ray are typically detected by the LYSO crystal.

The search for EC transition of ^{176}Lu can proceed by rejecting a large fraction of ^{176}Lu β-decay background by selecting an Energy_{LYSO} window. This analysis procedure and results are summarized in [11]. In the following a complementary analysis approach is adopted. A LYSO energy distribution that is abundant in EC signals can be obtained by selecting E_{Ge} in the "Bulk" of the expected signal region [$82.1\text{keV}-1\sigma < E_{Ge} < 82.1\text{keV}+1\sigma$] (see FIG. 2). By construction we expect that 68.3% of the total EC signal events belong to this [81.4 keV - 82.8 keV] energy range. We compare this last LYSO energy distribution with the one obtained by selecting E_{Ge} in the two "Sidebands" [$82.1\text{keV}-2\sigma < E_{Ge} < 82.1\text{keV}-1\sigma$] and [$82.1\text{keV}+1\sigma < E_{Ge} < 82.1\text{keV}+2\sigma$]. By construction only 13.6% of EC signal events lies in each sideband. The difference of the LYSO energy distributions obtained with "Bulk" and "Sidebands" events, will still contains 41% of the total number of the EC signal events, however the background contribution will be strongly suppressed[2].

Figure 4 shows the energy distributions measured by the LYSO detector for events selected in the "Bulk" range and in the "Sidebands", and also the residuals of the difference; no evidence for a statistically significant excess

[2]A complete background rejection occurs when the energy distribution of the background is a linear function in this (narrow) region.

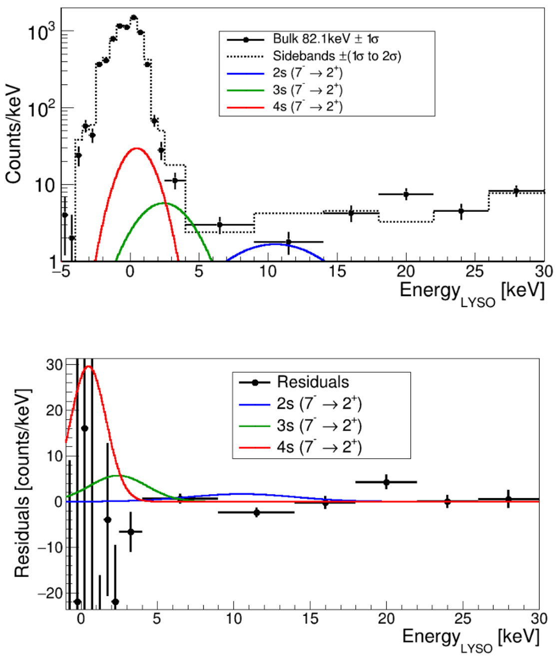

Figure 4. Top panel: Energy distributions measured by the LYSO detector for events selected in the [81.4keV< E_{Ge} < 82.8keV] range (±1σ "Bulk", data points) and in the two "Sidebands" (dashed lines). Bottom: residuals of the "Bulk"-"Sidebands" difference. The red, green and blue lines are the upper limits for the possible contribution of EC of the 4s, 3s and 2s electron of ^{176}Lu, respectively.

was found. Red, green and blue distributions shown in FIG. 4 are the 95% upper limits for the possible contribution of the Electron Capture of the 4s, 3s and 2s electrons, respectively; in particular the energy is emitted in the atomic rearrangement of the electron vacancy of the daughter, Yb, atom. Taking into account the 41% efficiency of this approach, the upper limits on the number of the ^{176}Yb$^{2^+}$ 82.1keV de-excitation γ-rays, N_{82}, are 210; 65 and 37 for EC of 4s; 3s and 2s electrons, respectively.

Following the approach of [5] an upper limit on the EC branching fraction can be obtained by comparing the upper limit on N_{82} with the number of events related with the β-decay branching fraction, thus evaluating the number of ^{176}Hf* events in the 88.3keV γ-peak in the HP-Ge event distribution (N$_{88}$=119.9 ± 0.5 × 10^3 events):

$$B_{82} = \frac{(1+\alpha_T^{82})N_{82}/\varepsilon_{88}}{(1+\alpha_T^{88})N_{88}} \quad (1)$$

where α_T^{82}=7.06 and α_T^{88}=5.86 are the total electron conversion coefficients for first excited levels of ^{176}Yb and ^{176}Hf, respectively [7], and $\varepsilon_{88}(82) \simeq 80\%$ is the detection efficiency loss of 82keV γ-ray with respect to 88keV γ-ray evaluated by Geant4 simulation and verified in this experiment by comparing the measured K$_\alpha$ and K$_\beta$ peak amplitudes of Lu and Hf with the 88keV peak [11].

On the other hand, with a probability 18.1% the 82.1 keV level can decay with a K-shell internal conversion [23] and with a probability 15.1% a 59.3 keV K$_\beta$ x-ray is emitted [1]. This is a different channel to search for the same EC decay to ^{176}Yb$^{2^+}$, providing additional (22.8 keV) energy released in the LYSO (see FIG. 5).

Table 1 is summarizing the upper limits obtained in the EC branching fraction by applying this analysis approach to the different decay channels. The expected distribution for the REC was evaluated by considering the approximations of [24] and assuming for ^{176}Lu a negligible contribution of Detour Transitions[3] [20, 21].

2.2 Search for ^{176}Lu EC to ^{176}Yb$^{0^+}$

The EC decay of ^{176}Lu to the fundamental state of ^{176}Yb is a 7th degree forbidden transition, thus it is expected to be suppressed with respect to the 5th degree forbidden EC decay in ^{176}Yb*; however if a ^{176}Lu nucleus captures a K-shell electron, the result must be a ^{176}Yb in the ground state. Considering that the K-shell atomic orbital is the one with the largest superposition with the nucleus, this possibility deserves a dedicated investigation.

A signature of the K-shell capture is provided by the x-rays emitted when the Yb atomic vacancy is filled. In particular, K$_{\alpha 2}$=51.35keV, K$_{\alpha 1}$=52.4keV and K$_\beta$=59.3keV are be emitted with a probability of 27.2%, 48.1% and 15.1%, respectively [1]. By comparing these x-ray energies with the measured energy distribution (see e.g. FIG. 3) it is clear that the Yb K$_\beta$ line is the only one distant enough from the nearby Lu and Hf K-shell lines, to be easily identified. Moreover, due to self absorption in the LYSO crystal and in the HP-Ge dead layers, the expected detection efficiency for Yb K$_\alpha$ lines is quite small, whereas the Yb K$_\beta$ line has $\varepsilon_{88} \simeq$100% [11].

Figure 5 shows the Energy distributions measured by the LYSO detector for events selected in the "Bulk" range of Yb K$_\beta$ and in the relative "Sidebands", and also the residuals of the difference; no evidence for a statistically significant excess was found. Thus, following the approach previously described, the upper limits on the number of the Yb K$_\beta$ 59.3keV x-rays, N_{59}, are 90 and 50 for the possible contribution of EC or REC of the 1s electron of ^{176}Lu, respectively. The upper limit to the corresponding branching fraction is:

$$B_{59} = \frac{N_{59}/(\varepsilon_{59}\varepsilon_{88})}{(1+\alpha_T^{88})N_{88}} \quad (2)$$

where ε_{59}=15.1% is the probability of a K$_\beta$ x-ray emission filling an Yb K-shell vacancy and $\varepsilon_{88} \simeq 100\%$ is the detection efficiency of 59keV x-ray with respect to 88keV.

3 Discussion and conclusions

The search for Electron Capture in ^{176}Lu is an opportunity for a laboratory measurement of 5th degree forbidden processes that may provide valuable information for nuclear theory models. Our active source technique, that uses a LYSO crystal scintillator also as detector, allows a powerful reduction of the background from ^{176}Lu β-decay and the HP-Ge intrinsic/external contamination.

[3] This cautiously minimizes the emitted energy in the REC signal distribution [11].

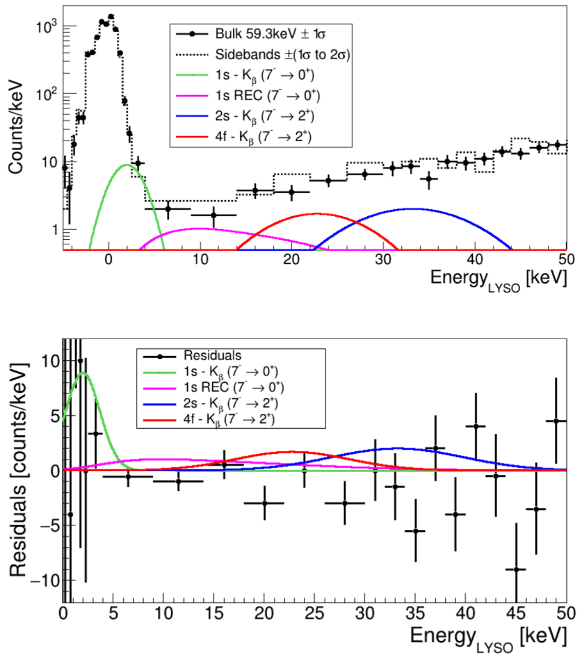

Figure 5. Top panel: Energy distributions measured by the LYSO detector for events selected in the [58.6keV< E_{Ge} < 60keV] range (±1σ "Bulk", data points) and in the two "Sidebands" (dashed lines). Bottom: residuals of the "Bulk"-"Sidebands" difference. The green and magenta lines are the upper limits for the possible contribution of EC or REC of the 1s electron of ^{176}Lu, whereas red and blue lines are the upper limits for the contribution of EC to ^{176}Yb$^{2^+}$ for 4f and 2s, respectively.

Table 1. Upper limits on the branching ratio (B.R.) for specific Electron Capture channels of ^{176}Lu.

EC/REC channel	**B.R. Limit (95% C.L.)**	**Previous Limit (68% C.L.) [5]**
1s EC + 59keV	0.07%	0.36%
1s REC + 59keV	0.04%	
2s/2p EC + 82keV	0.044%	0.45%
3s/3p EC + 82keV	0.083%	
3d EC + 82keV	0.11%	
4s EC + 82keV	0.26%	
4f EC + 59keV	0.26%	
2s REC + 82keV	0.06%	
2p REC + 82keV	0.05%	
3/4s REC + 82keV	0.055%	
3p REC + 82keV	0.075%	

No evidence for the EC process was found. Table 1 reports our estimation of the upper limits for the different EC channels. Improvements in sensitivity of 2-3 order of magnitude could be possible, in principle, by adopting the techniques of low-background experiments. In particular, by considering the possibility to operate small LYSO crystals as cryogenic bolometers, a very good signature of EC peaks in the LYSO spectrum is also expected [25].

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