

Measurement of the partial (n, γ) cross section to $^{176}\text{Lu}^m$ at s -process temperatures

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The s -process reaction path in the vicinity of ^{176}Lu has attracted considerable interest for a long time. Initially, ^{176}Lu was considered as a potential s -process chronometer due to its long half life of 36 Gyr. However, it turned out that the hot photon bath at typical s -process temperatures leads to thermally induced transitions between the long-lived ground state and the short-lived isomer (3.68 h), which are otherwise strictly forbidden. In this way the effective half life can be reduced to a few days. Accordingly, the final abundance ratio $^{176}\text{Hf}/^{176}\text{Lu}$ becomes a sensitive function of temperature and neutron density, which turns ^{176}Lu into an s -process thermometer. In this context, the isomeric ratio has to be known at the temperatures of the two neutron sources of the main s -process component, i.e. at 9×10^7 K ($kT = 8$ keV) and 2.7×10^8 K ($kT = 23$ keV), respectively. The stellar cross section feeding the isomeric state in ^{176}Lu has been measured at $kT = 5$ keV and 25 keV at the Karlsruhe 3.7 MV Van de Graaff accelerator via the activation technique. With these results and recent TOF data for the total capture cross section, the isomeric ratio was found to be constant in the relevant thermal energy range, in good agreement with a statistical model calculation. In the light of the improved cross section information, the branching at ^{176}Lu was analyzed with respect to the temperature conditions during He shell flashes in thermally pulsing low mass asymptotic giant branch stars.

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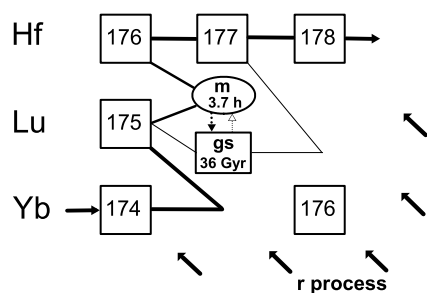


Figure 1: The s -process reaction flow in the Lu region. See text for the connecting arrows between $^{176}\text{Lu}^g$ and $^{176}\text{Lu}^m$.

1. Introduction

Because of their accurate solar abundances, the rare earth elements (REE) represent a reliable basis for detailed s -process analysis. Lutetium is the last REE and is followed in atomic number by the element hafnium. As shown in Fig. 1, the isobars ^{176}Lu and ^{176}Hf are shielded by ^{176}Yb against the r -process beta decay chains, and can, therefore, be considered to be of pure s -process origin. Due to its long half-life of 36 Gy, ^{176}Lu was initially considered as a potential nuclear chronometer for the age of the s elements. However, the thermal photon bath at typical s -process temperatures is energetic enough for induced transitions from the long-lived ground state to the short-lived isomer ($t_{1/2} = 3.68$ h), thus dramatically reducing the effective half-life to a few hours. This effect changes the information contained in the $^{176}\text{Lu}/^{176}\text{Hf}$ pair from a potential chronometer into a sensitive s -process thermometer (Beer et al. 1981; Klay et al. 1991; Doll et al. 1999).

As far as the stellar neutron capture rates are concerned, very accurate total (n, γ) cross sections have recently been obtained for ^{175}Lu and ^{176}Lu in a time-of-flight (TOF) measurement using a 4π BaF₂ array (Wisshak et al. 2006a). In case of ^{175}Lu it has to be considered that neutron captures may feed either the ground state or the isomer in ^{176}Lu . Therefore, the total (n, γ) cross section has to be complemented by a measurement of at least one of the two partial cross sections. Since the corresponding reaction channels could not be distinguished in the TOF measurement, the activation technique has been employed to determine the partial cross section to the isomeric state $^{176}\text{Lu}^m$ at thermal energies of $kT = 5$ and 25 keV.

2. Measurements

The activation measurements on ^{175}Lu were carried out at the Karlsruhe 3.7 MV pulsed Van de Graaff accelerator using two reactions for producing quasi-stellar neutron spectra close to the characteristic temperatures of the relevant neutron source reactions of the s process. Near the typical thermal energy of the $^{13}\text{C}(\alpha, n)^{16}\text{O}$ reaction at $kT = 8$ keV, a neutron spectrum corresponding to $kT = 5.1 \pm 0.1$ keV was obtained via the $^{18}\text{O}(p, n)^{18}\text{F}$ reaction by choosing the proton energy 8 keV above the reaction threshold [Heil et al. (2005)]. The neutron spectrum that can be produced via the $^7\text{Li}(p, n)^7\text{Be}$ reaction at $kT = 25$ keV represents the experimental counterpart of the second most important stellar neutron source provided by the $^{22}\text{Ne}(\alpha, n)^{25}\text{Mg}$ reaction at $kT = 23$ keV.

The Lu samples were cut from foils of natural composition. During the irradiations, each sample was sandwiched between two gold foils of the same diameter for normalization of the measured activation yield to the well known gold reference cross section. Since the proton energy is adjusted slightly above the reaction threshold, all neutrons are emitted in a forward cone of 140 deg opening angle.

After irradiation, the induced activities were measured with a γ -detection system consisting of two high purity germanium clover detectors. Each clover detector consists of four independent HPGe n-type crystals in a common cryostat. The whole assembly, which forms nearly a 4π geometry, was covered with 10 cm of lead in order to decrease the room background.

The analysis was based on the most intense line in the decay of the isomeric state with $E_\gamma = 88.36$ keV.

3. Analysis and results

Since all measurements are performed relative to ^{197}Au , the ratio between the number of activated nuclei in the sample and in the gold foils yields $\frac{A_i}{A_{\text{Au}}} = \frac{\sigma_i N_i f_{b_i}}{\sigma_{\text{Au}} N_{\text{Au}} f_{b_{\text{Au}}}}$ where A_i and A_{Au} are determined by the respective net counts registered in the detector, N is the number of sample atoms per cm^2 , σ the capture cross section, and f_b a correction factor that accounts for the decay and for the variation of the neutron flux during the irradiation.

Because of the low γ -energy emitted in the decay of $^{176}\text{Lu}^m$, considerable effort was devoted to the accurate determination of the corresponding self-absorption correction. The close geometry of the present experiment, which was necessary because of the weak activities produced in the 5 keV irradiations, required, however, that corrections were evaluated by comprehensive GEANT4 simulations. In these simulations minor effects due to the extended sample size and cascade summing have been included as well. The adopted Maxwellian averaged cross section (MACS) of gold at $kT = 5.1$ keV is $\frac{\langle \sigma v \rangle}{v_T} = 2028 \pm 50$ mb. [Heil et al. (2005)]

The weighted MACS for the partial (n, γ) cross section to the isomeric state of ^{176}Lu at $kT = 5.1$ keV is

$$\frac{\langle \sigma(^{176}\text{Lu}^m)v \rangle}{v_T} = 3048 \pm 195 \text{ mb.}$$

The uncertainty of the present measurement is clearly dominated by systematic effects, particularly by the intensity of the 88 keV line in the decay of $^{176}\text{Lu}^m$ and by the efficiency of the HPGe detectors. The problem with the uncertainty of the relative intensity was circumvented by comparison with the previous activations of Zhao and Käppeler (1991) at $kT = 25$ keV, where partial cross sections have been obtained by spectroscopy of the electrons emitted in the decay of $^{176}\text{Lu}^m$ as well as by detecting the 88 keV γ transition. The total capture cross section of ^{175}Lu has been accurately measured by Wisshak *et al.* (2006a). Combining their value at $kT = 25$ keV with the normalized partial cross section of 1153 ± 25 mb leads to an isomeric ratio of $IR(kT = 25 \text{ keV}) = 0.857 \pm 0.023$. At the lower thermal energy of $kT = 5$ keV the present measurement yields a partial cross section of 3185 ± 97 mbarn. The total capture cross section at $kT = 5$ keV was determined to be 3568 ± 75 mb following the prescription of Wisshak *et al.* (2006a). With these data, the isomeric ratio becomes $IR(kT = 5 \text{ keV}) = 0.893 \pm 0.037$.

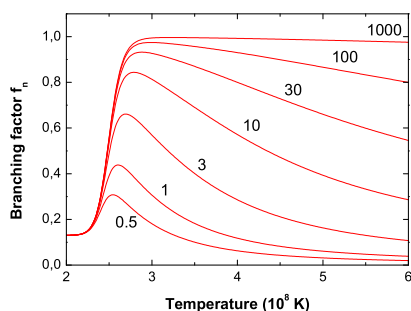


Figure 2: The branching factor f_n as a function of temperature and neutron density (in units of 10^8 cm^{-3}).

4. The ^{176}Lu branching and the s -process temperature

In addition to the experimental cross sections of ^{175}Lu reported here, MACSs of the other involved isotopes and the respective stellar enhancement factors were taken from Bao *et al.* (2000) and from the recent work of Wisshak *et al.* (2006a, 2006b).

The isomeric ratios in the keV region that are obtained by combining the total (n, γ) cross section data with the present results are significantly larger than at thermal neutron energy and practically constant. Within uncertainties, the present results at $kT = 5 \text{ keV}$ and 25 keV are in fair agreement. Though 8% higher, this behavior was expected by a theoretical prediction using the Hauser-Feshbach statistical model [Rauscher (2005)]. Therefore, a weighted average of 0.87 ± 0.03 has been adopted for the branching analysis. This result is compatible but more accurate than the value of 0.90 ± 0.04 derived on the basis of earlier cross sections [Bao *et al.* (2000)].

Contrary to the situation at low excitation energy, where transitions between ground state and isomer are strictly forbidden by selection rules, interaction with thermal photons induce transitions to higher lying nuclear states, which can decay into the ground state and into the isomer as well. This thermal coupling can, therefore, lead to a destruction of the long-lived ground state.

While thermal effects are negligible below $T_8 = 1.5$, where T_8 is the temperature in units of 10^8 K , the population probabilities of ground state and isomer start to be increasingly coupled at temperatures above $T_8 = 2.2$, resulting in a strong reduction of the effective half-life, but also in an additional feeding of the ground state. It is due to this effect that more ^{176}Lu is observed in nature than would be created in a "cool" environment. In this regime, internal transitions, β -decays, and neutron captures are equally important and have to be properly considered during He shell flashes, where the final abundance pattern of the s -process branchings are formed.

These thermal effects were treated by using the temperature-dependent beta decay rate of ^{176}Lu from the work of Doll *et al.* (1999) for calculating the branching factor as a function of temperature and neutron density (Fig.2), following the prescription of Klay *et al.* (1991) complemented by the updated input from Doll *et al.* (1999). This information is most crucial for the proper treatment of the strong temperature gradient in the bottom layers of the He shell flashes. Accordingly, the convective region of the He shell flashes was subdivided into 20 zones and the ^{176}Lu production and decay was followed over these layers numerically.

The Lu/Hf abundance ratio is required as an additional input for the branching analysis. The

adopted value of 0.239 has been taken from the work of Blichert-Toft and Albarède (1997), which is confirmed by data from Bizzarro et al. (2003) and is in perfect agreement with the value in the widely used solar system abundance table of Anders and Grevesse (1989).

In the investigated stellar scenario [Gallino et al. (1998)] too much of ^{176}Hf and somewhat too little of $^{176}\text{Lu}^g$ is produced during the main neutron exposure provided by the $^{13}\text{C}(\alpha, n)^{16}\text{O}$ reaction, where temperatures are too low for causing a thermal depopulation of the isomer. As indicated in Fig. 2 this additional feeding of the long-lived ground state occurs at temperatures between $T_8 = 2.2$ and 3, leading to a strong increase in the production of $^{176}\text{Lu}^g$ during He shell flashes.

One finds that ^{176}Hf is almost completely destroyed at the beginning of the flash, when neutron density and temperature are highest. Under these conditions, ^{176}Hf is efficiently bypassed by the reaction fbw (Fig. 2), resulting in a corresponding production of ^{176}Lu . As temperature and neutron density decline with time, the branching towards ^{176}Hf is more and more restored, but the final abundance at the end of the He shell flash remains significantly lower than before.

Compared to a similar previous study [Arlandini et al. (1999)], the now available, improved cross section information appears to settle the ^{176}Lu puzzle. While the ^{176}Hf abundance was before overproduced by 10 - 20%, preliminary calculations yield $^{176}\text{Lu}^g$ and ^{176}Hf abundances (relative to solar system values) of 1.04 and 0.96, respectively. These results do not yet consider the decay of the produced $^{176}\text{Lu}^g$ prior to the formation of the solar system, a correction that would bring the two numbers into close agreement. A quantitative estimate of this aspect has to be evaluated in the context of a galactic chemical evolution model.

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