International Conference on Nuclear Data for Science and Technology 2007
DOI: 10.1051/ndata:07408

Inelastic neutron scattering cross section of ^{187}Os at 30 keV

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Abstract. The Re/Os cosmochronometer requires a precise determination of the neutron capture cross section of ^{187}Os at the stellar conditions of *s*-process nucleosynthesis, i.e., the effective stellar values for thermal energies between 8 and 25 keV. This implies a correction to the cross section measured in the laboratory, because at these temperatures most of the ^{187}Os nuclei are in the first exited state at 9.75 keV. In order to support the necessary Hauser-Feshbach calculations of the capture cross section for the excited states, it is important to consider the competition by the inelastic channels, including the so-called super-elastic scattering, where the neutron gains in energy. In this context, the inelastic scattering cross section of ^{187}Os has been measured using a pulsed neutron beam with an energy of 30 ± 9 keV (FWHM) and a repetition rate of 1 MHz. Neutrons were produced directly at the threshold of the ^{7}Li(p,n)^{7}Be reaction in order to take advantage of the sharp collimation from reaction kinematics. To compensate for the low neutron yield close to threshold, scattered neutrons were recorded with three efficient ^{6}Li-glass scintillators 11 cm in diameter, located at 90 and 120 deg with respect to the neutron beam. Data have been taken with isotopically enriched ^{187}Os and ^{188}Os, the latter for measuring the elastic component separately. The analysis of the data was performed by a complete Monte Carlo simulation of the experiment. The results for the inelastic cross section are compared to previous data and are discussed with respect to the stellar (n,γ) rates, which are instrumental for an evaluation of the Re/Os cosmochronometer.

1 Introduction

The decay of ^{187}Re ($T_{1/2}$ = 41.2 Gyr [1]) can be used to date the onset of heavy element nucleosynthesis and – by extrapolation – the age of the cosmos. The amount of ^{187}Os produced by ^{187}Re decay can be evaluated by subtracting the component expected from *s*-process systematics from its solar abundance. In fact, ^{187}Os can be generated only by ^{187}Re β-decay or by the slow (*s*) neutron capture process. The typical ingredients of *s*-process modeling are the neutron capture cross sections of the involved nuclei. According to the classical approach the product of the abundance produced and the respective stellar cross section is constant for nuclei far from magic neutron numbers. With this relation the *s* component of ^{187}Os can be inferred from its neighbor isotope ^{186}Os, which is of pure *s*-process origin, simply on the basis of their stellar (n,γ) cross sections.

Apart from astrophysical considerations concerning the galactic chemical evolution [2–4], the largest nuclear uncertainty of the Re/Os cosmochronometer is due to the effective neutron capture cross section of ^{187}Os at the conditions of *s*-process nucleosynthesis. The fact that low lying excited states in ^{187}Os are more strongly populated than the ground state requires theoretical corrections by means of statistical model calculations for the (n,γ) cross sections of these states. In these calculations, good accuracy can only be achieved if the relevant input parameters can be extracted from experimental data. While level densities and strength functions are derived from the analysis of resolved resonances [5], the inelastic scattering cross section is important for describing the competition between the different reaction channels at stellar temperatures.

Besides from the problem of the (n,γ) cross section of ^{187}Os previous experimental data for ^{186}Os [6,7] differ especially at energies close to the first excited level (137 keV). The discrepancies in the ratio of the stellar Maxwellian averaged cross sections of these two isotopes reported by the different measurements and calculations [8,9] correspond to an uncertainty of 2.3 Gyr for the estimated age. In order to reduce these uncertainties, the neutron capture cross sections of ^{186}Os and ^{187}Os have been measured and analyzed from 1 eV to 1 MeV at the n_TOF spallation neutron source [5,10]. In addition to this study, the inelastic scattering cross section has been investigated at the 3.7 MV Van de Graaff accelerator of Forschungszentrum Karlsruhe. So far, the inelastic scattering cross section for the first excited level of ^{187}Os has been measured at 24, 34, 45, and 60.5 keV neutron energy with quoted uncertainties between 13 and 30% ([11–14], values plotted in fig. 5). The extrapolation of the transmission functions for all inelastic channels at stellar conditions would greatly benefit from an improvement of this cross section, in particular by an additional accurate measurement at astrophysical energies.

2 Measurement techniques

The aim of the present measurement is to discriminate elastically and inelastically scattered neutrons in a time-of-flight (TOF) experiment. In order to use their difference in energy, a sufficiently narrow neutron energy distribution is required, e.g., comparable or less than the energy difference defined by the 9.75 keV of the first excited level. Application of the TOF method is attractive because three over the four available

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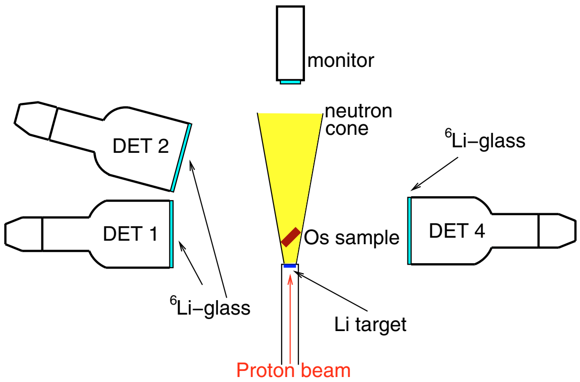

Fig. 1. Sketch of the experimental setup (not to scale). The sample (15 mm in diameter and 1.6 mm in thickness) is mounted at a distance of 4 cm from the lithium target. The flight paths from the sample to the detectors are 26.1 cm and the neutron monitor is located 1.11 m from the target.

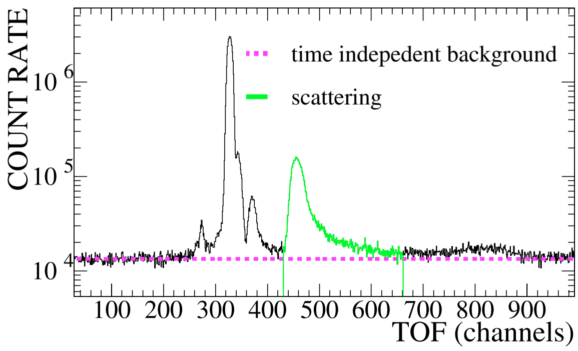

Fig. 2. Raw TOF data of ^{187}Os taken with DET 1. The linear fit of the count rate to the left of the peaks has been used to define the time-independent background. The first peak from the left is due to (p,γ) interactions in the slits of the analyzing magnet close to the lithium target. The second peak represents the γ-flash when the proton pulse hits the target; this peak overlaps with γ-rays from neutron captures in the sample. The next peak before the distribution produced by scattered neutrons stems from the 5.9 keV (n,γ) resonance in the aluminum of the sample canning, and the feature at channel 800 is due to (n,γ) reactions in the monitor.

measurements (at 24, 34, 45 keV) were performed by means of neutron filters, using deep interference minima in the cross sections of different materials [11–13], which allows one to produce quasi monoenergetic neutrons with an energy resolution of several keV from an originally continuous spectrum. Only one of the previous measurements was performed with a truly monoenergetic neutron beam [14]. This technique has the advantage to have a better identifiable elastic component and a cleaner background, although it is difficult to produce a narrow monochromatic neutron beam below 100 keV. The ^{7}Li(p,n)^{7}Be reaction is best suited for its comparably large neutron yield at 30 keV.

3 Experimental setup

The present measurement used the ^{7}Li(p,n)^{7}Be reaction at the threshold of 1.8811 MeV. A narrow neutron spectrum centered at 30 keV was obtained by keeping the proton energy within a range of 0.7 keV above the threshold. Since this was below the energy spread of the beam, part of the beam intensity of 2 μA up to 6 μA had to be sacrificed in order to get a sufficiently small FWHM for the neutron energy distribution.

The targets consisted of natural metallic lithium layers 1 μm in thickness, which were evaporated onto silver backings to minimize background from energetic γ rays due to (p,γ) reactions. The neutron energy was measured via TOF by operating the accelerator in pulsed mode with a frequency of 1 MHz and a pulse width of 10 ns. The resulting energy distribution has been found to be essentially of Gaussian shape with a FWHM between 7.6 keV and 9.5 keV depending on the actual proton energy. Since this spread was slowly drifting between these limits with a time constant of several hours, the energy distribution was constantly monitored. In this way, it was possible to select runs with similar energy resolution.

At the reaction threshold, neutrons are emitted in a narrow cone in forward direction. Under the conditions of the present measurements the maximum emission angle was always below 14 deg. Neutrons were detected with ^{6}Li-glass scintillators. The monitor, which consisted of GS20 (NE908) 3 cm in diameter and 3 mm in thickness, was placed at a distance of 1.11 m from the lithium target. Scattered neutrons were recorded with three KG2 (NE913) ^{6}Li-glass detectors [15] 11 cm in diameter and 3 mm in thickness. The scintillators, mounted on 9823QKA Thorn EMI photomultiplier tubes, were placed outside the neutron cone at a distance of 26.1 cm from the sample at angles of 90 (DET1), 120 (DET2), and 270 deg (DET4) with respect to the beam axis (fig. 1).

The lithium target was surrounded by a lead ring in order reduce the γ-background from the target in DET1 and DET4. The ^{187}Os and ^{188}Os samples used in the experiment consisted of isotopically enriched metal powder loaned from Oak Ridge National Laboratory. The samples were encapsulated in thin aluminum cans 15 mm in diameter and ~1.6 mm in thickness and glued to a KAPTON© foil sustained by a carbon fiber frame outside the neutron cone. As indicated in figure 1 the samples were mounted at 45 deg at a distance of 4 cm from the lithium target.

The ^{188}Os sample was included in the experiment for obtaining the spectrum from elastic scattering and an empty was used to characterize the ambient background. The data acquisition system was based on Flash Analog-to-Digital Converters (FADC) to digitize the detector signals with a sampling rate of 1 ns/channel.

4 Data analysis and preliminary results

The digitized data recorded with the FADC allowed us to implement a pulse shape analysis of every signal and also to perform a fast check of the neutron energy distribution.

During the entire experiment the neutron energy spectrum was continuously analyzed in time intervals from a few seconds to a few minutes depending on the counting statistics in the monitor detector. For every event the TOF, the integral over the signal, and the ratio between the area of the first 10 ns of the signal and the total integral were recorded. The latter information was used for a simple pulse shape discrimination to reduce the γ-background. In general,

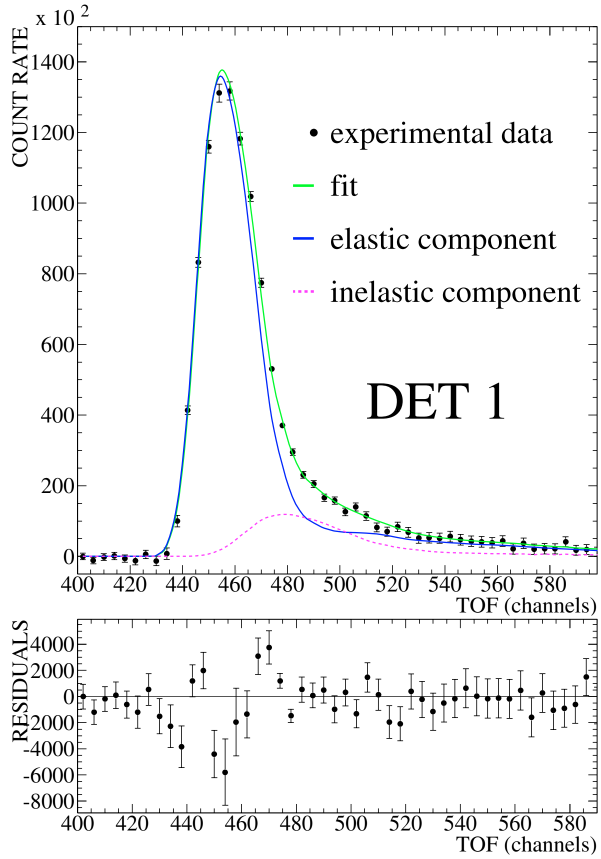

Fig. 3. Top: The background subtracted TOF spectrum of ^{187}Os taken with DET 1 and a fit to the data (reduced χ^2 = 1.4). Bottom: The residuals of the fit. The errors bars are mainly due to the systematic uncertainties resulting from background subtraction.

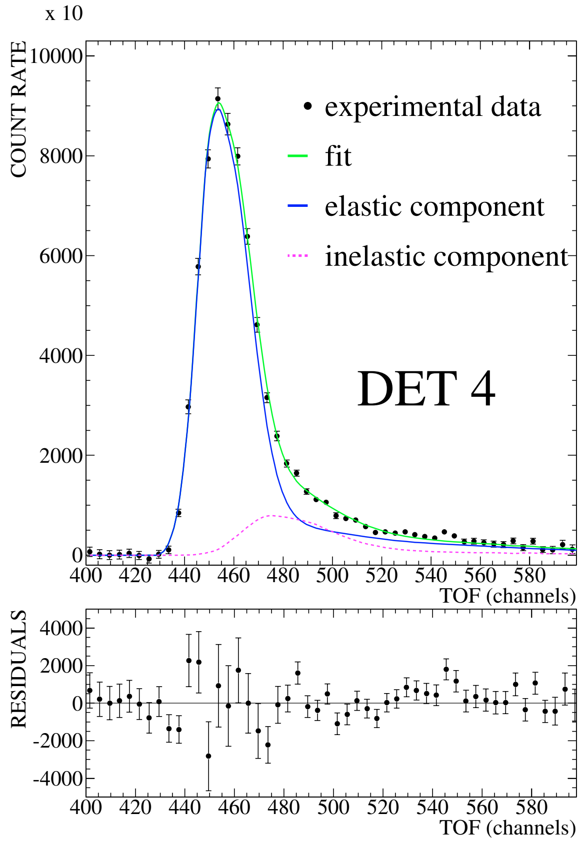

Fig. 4. Top: The background subtracted TOF spectrum of ^{187}Os taken with DET 4 and the corresponding fit (reduced χ^2 = 1.4). Bottom: The error bars of the residuals are mainly due to systematic uncertainties resulting from the background subtraction.

the detection of neutrons, which corresponds to an energy deposition of 4.8 MeV from the ^{6}Li(n,α)^{3}H reaction, produces a characteristic signal with a slower decay than the shorter shapes due to γ-rays.

Though the pulse shape analysis provides not a perfect separation between γ-rays and neutrons, the partial reduction was significantly improving the signal to background ratio. The remaining background components in the TOF spectra were:

1. the γ-flash generated by the impact of the proton pulse on the target (well separated from events due to scattered neutrons),
2. the (n,γ) events in the sample and (at larger TOF) in the monitor,
3. a constant time-independent background defined by the events prior to the γ-flash.

All these backgrounds could be clearly identified (see fig. 2).

The time-independent background is affecting the scattering distribution the most and requires a correction of about 10%. Most background components listed above can derived from the measurement with the empty sample canning. The elastic scattering distribution from the count rate of the ^{188}Os sample and the elastic plus inelastic distribution measured with the ^{187}Os sample are recorded with statistical uncertainties smaller than 1%. By combining runs with similar FWHM in neutron energy, comparison of the yields from the two samples yields a robust indication for the inelastic part, although the separation from the elastic peak is not clearly visible (see also ref. [10]).

The spectra recorded by the detector at 120 deg (DET2) exhibit the signature of moderation due the glue used in closing the sample cans. Because the elastic scattering cross section of hydrogen is strongly angle dependent and favors forward angles, these neutrons arrive at the detector with a delay and appear therefore as a bump in the tail of the scattering distribution.

In order to evaluate this effect properly, detailed GEANT-MICAP simulations of the complete experiment have been performed. The simulations included the neutron energy spectrum according to the kinematics of the ^{7}Li(p,n)^{7}Be reaction. In this way, the components in the TOF spectra due to scattering in the moderator and in the aluminum can were separated by the one in osmium. The so obtained TOF distribution generated by the moderation effect were matched to the visible effect in DET2. For the other two detectors, which were much less affected, this component was scaled accordingly. The simulated background components due to moderation have been checked to fit the level estimated from the measurement with the empty can. For all the detectors, it has been found to be not larger than 10% of the scattering distribution.

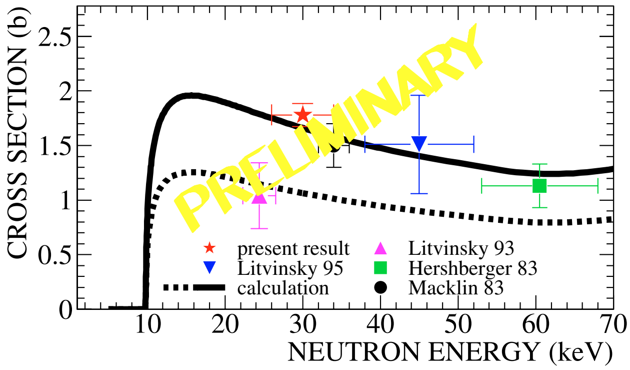

Fig. 5. Inelastic scattering cross sections from refs. [11–14] compared to the present preliminary result obtained by means of the ratio of elastic and inelastic components derived from the present measurement. The elastic cross section was adopted from refs. [11, 13]. The dotted line represents a Hauser Feshbach calculation performed without any experimental input for the inelastic parameters, the solid line is the same calculation rescaled to match the weighted average of all the experimental values.

After subtraction of all background components, the spectra have been fitted using the TOF distributions obtained in the simulations. It was found that the simulated elastic spectra were matching the measured ^{188}Os spectrum with a reduced χ^2 of 1.1 and 1.4 for the detectors at 270 deg (DET4) and 90 (DET1), respectively. The only free parameter in this fit was the normalization of the number of events in the TOF distribution.

The inelastic component was then constructed by shifting the TOF distribution of the elastic component by the energy transfer for exciting the first level in ^{187}Os at 9.75 keV. The only free parameters in the corresponding fit were, therefore, the number of events due to elastic and inelastic scattering in the ^{187}Os spectrum.

The fits of the scattering spectra measured with a neutron energy distribution of 9.5 keV FWHM are shown in figures 3 and 4. The ratio elastic/inelastic has been determined with an uncertainty of about $\leq 8\%$. Our preliminary result is in good agreement with the measurements reported in refs. [12–14]. From the ratio, a first guess for the inelastic scattering cross section has been obtained using previous elastic cross section data (fig. 5). The final inelastic scattering cross section value will be determined using the elastic scattering cross section of ^{188}Os obtained by the total cross section minus the neutron capture cross section measured at n_TOF for normalization.

5 Conclusions

The inelastic scattering cross section of ^{187}Os was successfully measured directly at the threshold of the ^{7}Li(p,n)^{7}Be reaction in spite of the small neutron yield. The fairly clean experimental conditions reached in this experiment illustrate that the monoenergetic neutron beam of 30 keV can be used to determine inelastic scattering cross sections for low lying excited states. Using a simple exponential model for the *r*-process production of ^{187}Re and recently determined isotopic and elemental abundances [16], we found that the remaining uncertainties of the neutron capture rates of [10] and of the present result give rise to an uncertainty in the duration of nucleosynthesis of 3% or 0.5 Gyr for a reference age of 15 Gyr. This "nuclear cross section" uncertainty is smaller than the ones estimated from astration effects and chemical evolution modeling.

We are thankful to D. Roller, E.-P. Knaetsch, and W. Seith for their continuous support during the measurements.

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