Measurement of the $^7$Be$(n, p)$ cross section at thermal energy


1Nuclear Physics Institute, Academy of Sciences of the Czech Republic, CZ-25068 Řež, Czech Republic
2Institut Laue-Langevin, 71 av. Martyrs, F-38042 Grenoble 9, France
3Research Centre Rez, CZ-25068 Řež, Czech Republic
4Laboratory of Radiochemistry, Paul Scherrer Institut, CH-5232 Villigen, Switzerland
5ISOLDE, CERN, CH-1211 Geneva 23, Switzerland
6Institut für Physik, Johannes Gutenberg-Universität, Mainz, Germany
7School of Physics and Astronomy, The University of Manchester, Manchester, United Kingdom
8Petersburg Nuclear Physics Institute, NRC Kurchatov Institute, 188300 Gatchina, Russia
9Institute for Materials Science and Center for Nanointegration, Duisburg-Essen (CEINDE), University of Duisburg-Essen, 45141 Essen, Germany

(Received 7 September 2018; published 14 January 2019)

The $^7$Be$(n, p)$ cross section was measured with an ion-implanted $^7$Be target at a thermal neutron beam of the research reactor LVR-15 in Rež. The cross section to the ground state of $^7$Li is $\sigma(n, p_0) = 43800 \pm 1400$ b and the cross section to the first excited state of $^7$Li is $\sigma(n, p_1) = 520 \pm 260$ b.

DOI: 10.1103/PhysRevC.99.014612

I. INTRODUCTION

Using the precise measurement of baryon density in the universe via precision measurements of the cosmic microwave background (CMB), the standard theory of big bang nucleosynthesis (BBN) can provide a quite precise prediction of primordial abundances of the light isotopes [1]. These predictions can be compared with the experimentally observed abundances. It has been confirmed that the BBN predictions for abundances of primordial deuterium, $^3$He and $^4$He relative to hydrogen, are in a good agreement with experimentally observed values. The success of this theory, however, sharply contrasts with the failure of the BBN $^7$Li abundance prediction. The predicted $^7$Li abundance data exceeds the telescope observations of very old metal-poor halo stars, which are considered a good surrogate for the primordial abundance, by more than a factor of three [2]. The cause of this discrepancy, which became known as the cosmological $^7$Li problem, might be related to nuclear reaction rates that are not well known. Because the primordial $^7$Li is predominantly produced by electron capture decay of the primordial $^7$Be, the production and destruction reaction rates of $^7$Be are crucial parameters for the evaluation of the $^7$Li abundance. The reaction rate of the indirect $^7$Be destruction chain $^7$Be$(n, p)^7$Li$(\alpha, n)$ is one of these parameters that is still not well known. In this work, we measured the cross section of the $^7$Be$(n, p)^7$Li reaction at thermal energy (25.3 meV). This thermal neutron cross section serves as a benchmark for normalization of neutron capture data that correspond to primordial temperatures, i.e., in the neutron energy range of tens to hundreds of keV.

Despite the extremely high cross section of the $^7$Be$(n, p)^7$Li reaction, the measurement of this value with a required precision is not an easy task. It is mainly due to a relatively short half-life of the $^7$Be target ($T_{1/2} \approx 53.2$ d) and the resulting high-$\gamma$ activity of such targets. Moreover, the preparation of high-quality $^7$Be targets, i.e., with a well-defined size, thickness and homogeneity, is not straightforward.

The first measurement of the $^7$Be$(n, p)$ thermal cross section was carried out by Hanna [3] already in 1955. For thermal neutron capture, the $^2^+$ capture state decays dominantly by proton emission. Thus, the obtained value of the $(n, p)$ reaction, $\sigma_{np} = 53000 \pm 8000$ b, can be directly compared with the total thermal neutron capture destruction cross section, $\sigma_{tot} = 48000 \pm 9000$ b, given in the same paper. Other measurements at thermal neutron energy were performed by Gledenov et al. [4] and Cervena et al. [5], both obtained $\sigma_{np}$ values in accordance with the measurement of Hanna et al., i.e., $50000 \pm 1000$ b and $46800 \pm 4000$ b, respectively. A significantly different cross section of $\sigma_{np}$ was, however, measured by Koehler et al. [6] in Los Alamos: $38400 \pm 200$ b for the transition to the ground state of $^7$Li($p_0$) and 420 $\pm 120$ b for the transition to the first excited state of $^7$Li ($p_1$). These data were obtained with a relatively high precision (2% for the $p_0$ transition, 29% for the $p_1$ transition). However, to date...
it has not yet been elucidated why $\sigma_{np}$ measured by Koehler et al. is significantly smaller (by about 15–20%) than the values measured by other groups.

Our current experiment and the recent experiments at the CERN nTOF facility [7,8] were performed as part of a systematic investigation of all neutron-induced destruction reactions of $^7$Be [9] at various neutron energies up to primordial temperatures.

II. EXPERIMENTAL DETAILS

The measurement of the $^7$Be($n$, $p$) cross section at thermal energy was performed at the LVR-15 research reactor operated by Research Centre Rez. The pure thermal neutron beam from the 6-m long neutron guide [10] was exploited for the experiment. The thermal neutron equivalent flux of the beam is $10^7$ cm$^{-2}$ s$^{-1}$, with the cadmium ratio of $10^5$. The neutron beam was trimmed by the neutron guide to a height of 4 mm and a width of 40 mm. The beam intensity and fluence were monitored by a monitor system placed at the neutron guide exit via the $^6$Li($n$, $\alpha$) reaction measured with a charged particle detector in a monitor chamber.

The $^7$Be target was fabricated using $^7$Be chemically extracted from the cooling water of the SINQ facility at PSI [11]. This $^7$Be sample was then introduced into an ISOLDE target and ion source unit and shipped to CERN. There the sample was heated, the released $^7$Be atoms were resonantly laser ionized and accelerated to 30 keV. After mass separation the $^7$Be ion beam was focused loss free through a 5 mm diameter diaphragm and implanted into a 1.5 $\mu$m thin Al foil [11]. The resulting beam spot had about 3 mm diameter. This Al foil was mounted between two 1.5 mm thick Al frames, with the external dimensions of $112 \times 50.5$ mm$^2$. The accurate position of the $^7$Be spot with respect to the Al frame was examined by an autoradiographic measurement using a flat panel detector imaging plate. The number of the $^7$Be atoms in the implanted spot was determined via $\gamma$-ray spectrometry measurements of the sample. A high-purity Ge detector with relative efficiency of 18% (relative to a 3 in. diameter by 3 in. long NaI crystal at 25 cm distance and specified at 1.332 MeV) and FWHM = 1.8 keV for energy of 1332 keV was used for the activity measurement. The detector was placed in a shielding box with 5 cm thick lead walls. The point radionuclide standards of EG3 type, $^{60}$Co, $^{133}$Ba, $^{137}$Cs, $^{241}$Am, and $^{152}$Eu, from Czech metrology institute were utilized for the energy and efficiency calibration. The certificated total error of activities for all these standards does not exceed 1%. The relative high activity of $^7$Be sample allowed using the detector-source distance of 75 cm, thus uncertainties connected with geometry factors, dead time correction, and random coincidences and statistical error were optimized. The activities of the $^7$Be target at the beginning of the two $^7$Be($n$, $p$)Li measurements were found to be $(16.46 \pm 0.32)$ MBq and $(8.57 \pm 0.17)$ MBq, the effective number of the $^7$Be atoms in the target, was derived from the activity using a half-life of $T_{1/2} = 53.23 \pm 0.04$ d. This value is the weighted average of $T_{1/2} = 53.17 \pm 0.07$ d [12] and $T_{1/2} = 53.257 \pm 0.044$ d [13], the two literature values reported for dilute $^7$Be atoms embedded in aluminium. The decay rate of $^7$Be is very slightly host dependent, therefore we used this weighted average value for $^7$Be in Al rather than the evaluated value [14].

For determination of the $^7$Be($n$, $p$) cross-section, a comparative method with a high-quality $^6$LiF standard, supplied by the Institute for Reference Materials Measurements (IRMM) Geel, was used. The standard sample was prepared by magnetron sputtering as a $^6$LiF homogeneous layer with a thickness of 30.2(2) $\mu$m/cm$^2$ deposited on a 1.5 $\mu$m Al foil. The diameter of the standard was 30 mm, the homogeneity of it was examined by the coincidence technique using two Timepix detectors [17]. In order to keep the geometry for both $^6$LiF and $^7$Be samples similar (that would optimize statistical and systematic uncertainties) and to fit our neutron beam, the measured area of the $^6$LiF standard was shrunk. To do that paper diaphragms of thickness of 250 $\mu$m were used to form an aperture that was placed on the $^6$Li sample surface. The photo of this standard is depicted in Fig. 1. For the main experiment an aperture with an 11 mm diameter was used. This diameter optimizes combined uncertainties associated with the geometry of the experiment and the number of Li atoms not covered by the diaphragm. Together with the diaphragms, the $^6$LiF standard was fixed on a plastic frame with the same size as the Al frame, and altogether it was placed at the same position as the Al frame with the $^7$Be sample.

The $^7$Be sample and the $^6$LiF standard were irradiated with a collimated thermal neutron beam in a large vacuum chamber. For the measurements the $^7$Be and $^6$LiF targets were fixed to a special holder that was constructed to match exactly the external dimensions of the Al or plastic frames. This enabled, unambiguously, to reproduce the same position for both targets. During the measurement the targets were tilted by 15° with respect to the neutron beam plane. This enabled to avoid self-shielding of the neutron beam and irradiate the whole measuring area on the targets. The charged particle spectra were taken with a fully depleted surface barrier detector with a 300 $\mu$m sensitive depth and 50 mm$^2$ active area. The position

FIG. 1. The LiF standard with the paper diaphragm upon the plastic frame.
TABLE I. List of reaction products, their original energies and their final energies after passage through 5.2 \( \mu \text{m} \) Al foil calculated with SRIM code [16].

<table>
<thead>
<tr>
<th>reaction</th>
<th>original energy (MeV)</th>
<th>final energy (MeV)</th>
<th>original energy (MeV)</th>
<th>final energy (MeV)</th>
</tr>
</thead>
<tbody>
<tr>
<td>(^7\text{Be}(n, p)^0\text{Li}_0 )</td>
<td>1.44</td>
<td>1.23</td>
<td>0.21</td>
<td>DNT</td>
</tr>
<tr>
<td>(^7\text{Be}(n, p)^1\text{Li}_1 )</td>
<td>1.02</td>
<td>0.76</td>
<td>0.15</td>
<td>DNT</td>
</tr>
<tr>
<td>(^6\text{Li}(n, \alpha)^t\text{t} )</td>
<td>2.05</td>
<td>0.43</td>
<td>2.73</td>
<td>2.40</td>
</tr>
<tr>
<td>(^{10}\text{B}(n, \alpha)^0\text{Li}_0 )</td>
<td>1.47</td>
<td>DNT</td>
<td>0.84</td>
<td>DNT</td>
</tr>
<tr>
<td>(^{10}\text{B}(n, \alpha)^1\text{Li}_1 )</td>
<td>1.77</td>
<td>0.11</td>
<td>1.01</td>
<td>DNT</td>
</tr>
</tbody>
</table>

1DNT: does not transmit through 5.2 \( \mu \text{m} \) Al foil

of the detector was perpendicular towards the target plane at a distance of 36 mm.

In our previous measurements with the \(^7\text{Be} \) sample it was found that the aluminium foil used to implant the \(^7\text{Be} \) also contained trace amounts of \(^{10}\text{B} \). The \( \alpha \) particles from the \(^{10}\text{B}(n, \alpha)^7\text{Li} \) reaction could cause an unwanted interference with protons from the \(^7\text{Be}(n, p) \) reaction of interest. To avoid this interference, a thin Al foil with a thickness of 5.2 \( \mu \text{m} \) was inserted in front of the detector. The foil enabled to stop these \( \alpha \) particles, but protons could pass through the foil though with a small energy loss, see SRIM [16] calculation in Table I.

In the main experiment, an eight-day measurement with \(^7\text{Be} \) (with 8.57 \( \pm \) 0.17 MBq activity) and a three-day measurement with \(^6\text{LiF} \) (with a delimited area of 11 mm in diameter) were combined. The results of these measurements are described in detail in the following section. At the end of this section we compare and combine this main experiment with two prior measurements obtained with the Li standard delimited to circular areas of 3 mm and 20 mm diameter.

III. RESULTS AND DISCUSSION

The spectrum of the charged particles for the 8.57 MBq \(^7\text{Be} \) sample taken with the thin fully depleted detector is depicted in Fig. 2. As mentioned above, the 5.2 \( \mu \text{m} \) Al foil was placed between the target and the detector. It enabled to remove unwanted reaction products from the spectrum. Thus, the \( \alpha \) background from the \(^{10}\text{B}(n, \alpha)^7\text{Li} \) and \(^{6}\text{Li}(n, \alpha)^t\text{t} \) reactions (Li trace contamination was also found in the \(^7\text{Be} \) target) was removed, and a possible interference between the 1.44 MeV \( p_0 \) (from \(^7\text{Be} \)) and 1.47 MeV \( \alpha_0 \) (from \(^{10}\text{B} \)) particles was eliminated. In Fig. 2, however, other peaks \( p(\text{14N}) \) and \( t(\text{6Li}) \), besides \( p_0 \) \(^7\text{Be} \) and \( p_1 \)^7\text{Be}, can be seen. They originate from the \((n, p)\) and \((n, t)\) reactions on the \(^{14}\text{N} \) and \(^6\text{Li} \) contaminants, respectively, that could not be suppressed. Nitrogen contribution is caused by residual air in vacuum chamber, roughly \( 10^{-3} \) of normal pressure. Minor contamination of construction material is responsible for lithium peak in the \(^7\text{Be} \) spectrum. A similar peak can also be observed in the spectrum for the Li standard.

The 5.2 \( \mu \text{m} \) Al filter affects the energy of protons, though only slightly (from a SRIM simulation [15,16] the energy loss for 1.44 MeV \( p_0 \) was found to be about 200 keV). The exponentially rising low-energy background is mainly due to the \( \beta' \)s (with energies up to 2.9 MeV) from decay of \(^{28}\text{Al} \) produced by neutron activation of the Al foil.

The \(^6\text{LiF} \) standard was irradiated in the same geometry as the \(^7\text{Be} \) sample, including the 5.2 \( \mu \text{m} \) Al filter. The spectrum from the three-day measurement is shown in Fig. 3. This
The charged particle spectrum of the LiF standard. Note that the labels on the top axis represent only a rough energy calibration.

The spectrum corresponds to the tritons with the dominant full energy peak Li(t). A small background peak Li\textsubscript{back}(t) caused by tritons (from \textsuperscript{6}Li) that did not pass through the Al filter can be identified in the vicinity of Li(t). By comparison of both spectra (Figs. 2 and 3), it is evident that the unwanted contribution of the background caused by Li\textsubscript{back}(t) tritons can be neglected.

Both measurements were normalized using a neutron-beam monitor. The monitor comprised a thin target (\textsuperscript{6}Li implanted into a Si wafer), inserted into the neutron beam, and a 50 mm\textsuperscript{2} PIPS detector (out of the beam) that was separated from the target by a 10 mm distance. The signals from the monitor were processed by a standard spectroscopic chain. For monitoring of the neutron beam, only 2.73 MeV tritons were registered and counted. The monitor quantified the total neutron beam fluence for all measurements. Considering the same detection efficiency (for both 2.73 MeV tritons and 1.4 MeV protons, and the same sample-detector distance for both the \textsuperscript{7}Be sample and the \textsuperscript{6}LiF standard, and taking into account a perfect 1/\nu dependence of the neutron cross sections for both \textsuperscript{6}Li [18] and \textsuperscript{7}Be [6] up to 100 keV, the cross section of the \textsuperscript{7}Be(n, p\textsubscript{0}) reaction at thermal energy (0.0253 eV), \(\sigma[\textsuperscript{7}Be(n, p\textsubscript{0})]\), could be determined via the following equation:

\[
\sigma[\textsuperscript{7}Be(n, p\textsubscript{0})] = \sigma(\textsuperscript{6}Li) \frac{A(p\textsubscript{0})}{A(Li)} \frac{M_{\textsuperscript{6}Li}}{M_{\textsuperscript{7}Be}} \frac{N(\textsuperscript{6}Li)}{N(\textsuperscript{7}Be)} C. \quad (1)
\]

Here, \(\sigma(\textsuperscript{6}Li)\) is the neutron cross section of the \(\textsuperscript{6}Li(n, \alpha)\)t reaction for the 25.3 MeV neutrons, \(A(p\textsubscript{0})\) and \(A(Li)\) are areas of the \(p\textsubscript{0}\) and \(t\) peaks in the \(\textsuperscript{7}Be(n, p\textsubscript{0})\) and \(\textsuperscript{6}Li(n, \alpha)\)t spectra, respectively, \(M_{\textsuperscript{6}Li}\) and \(M_{\textsuperscript{7}Be}\) are neutron flux monitor values for the \(\textsuperscript{7}Be(n, p\textsubscript{0})\) and \(\textsuperscript{6}Li(n, \alpha)\)t measurements, respectively, \(N(\textsuperscript{6}Li)\) and \(N(\textsuperscript{7}Be)\) are numbers of \(\textsuperscript{6}Li\) atoms in the delimited area of the \(\textsuperscript{6}LiF\) standard and an effective number of the \(\textsuperscript{7}Be\) atoms in the sample, respectively, and \(C\) is a geometry correction factor.

The effective number of the \(\textsuperscript{7}Be\) atoms, \(N(\textsuperscript{7}Be)\), was derived from the activity of the sample. To evaluate \(N(\textsuperscript{7}Be)\), an initial activity [i.e., 8.57(17) MBq] and a drop off of the \(\textsuperscript{7}Be\) atoms (due to the radioactive decay) throughout the measurement were taken into account. The correction and uncertainty associated with a time variation of neutron flux can be neglected (<0.1%). Considering the stability and uniformity of the \(\textsuperscript{6}LiF\) standard, the number of the \(\textsuperscript{6}Li\) atoms in Eq. (1), \(N(\textsuperscript{6}Li)\), is given by a nominal density \(\textsuperscript{6}LiF\) film, 30.2(2) \(\mu\text{m/cm}^2\), and the diameter of a selected pin hole delimiting the measured area, i.e., 11 mm. With respect to different effective areas of the \(\textsuperscript{7}Be\) sample and the \(\textsuperscript{6}LiF\) standard, the normalized area ratio in Eq. (1) has to be corrected for a different measurement solid angle and imperfect uniformity of the thermal neutron beam. In order to evaluate this correction, the neutron beam profile was measured by a gadolinium-doped imaging plate. Due to the construction of the neutron guide, the beam inhomogeneity was found to be noticeable mainly in the vertical direction. Using the digitized beam profile, the effective dimensions of the \(\textsuperscript{6}LiF\) standard and \(\textsuperscript{7}Be\) sample and geometry parameters, the correction was calculated to be 14.7(7)%.

To evaluate \(\sigma[\textsuperscript{7}Be(n, p\textsubscript{0})]\) according the Eq. (1), the value of \(\sigma(\textsuperscript{6}Li) = 940(4) \text{ b}\) for 25.3 MeV neutrons was adopted from the Neutron Cross Sections catalog [19]. It is based on the old but relatively precise measurements [20,21]. Inserting all variables into Eq. (1), the cross section of the \(\textsuperscript{7}Be(n, p\textsubscript{0})\) reaction at the thermal energy (0.0253 eV) was evaluated, \(\sigma[\textsuperscript{7}Be(n, p\textsubscript{0})] = 43500 \pm 700(\text{stat.}) \pm 1700(\text{syst.}) \text{ b}\).
The statistical error comes from the measured energy spectra. The systematic error incorporates uncertainties of the $^6$Li thermal neutron cross section, 0.4%, the nominal value of the $^6$LiF standard thickness, 0.7%, the delimited area of the standard, 3.7%, the $^7$Be activity, 2%, the distribution of the $^7$Be atoms within the sample spot, 0.8%, and uncertainties associated with the geometry and beam profile, 2.9%. The influence of the 5.2 μm Al filter on the effective solid angle of the transmitting particles was also examined. Both protons and tritons from the $^7$Be sample or $^6$LiF standard can, in principle, be deflected in the Al foil from their original direction. To evaluate this effect, the SRIM code [16] was employed. The simulation showed that the deflection of the proton and triton particles in the thin Al filter is negligible (the average deflection angle for 2.7 MeV tritons is 1.5° and the probability of missing the detector due to this deflection is below $10^{-4}$) and can be neglected.

Two additional comparative measurements with the $^6$LiF standard were performed using 3 mm and 20 mm apertures. These measurements were carried out in a compact geometry with the detector sample ($^7$Be or $^6$LiF, respectively) distance of 19 mm. For this arrangement, the Al filter was removed. In these experiments the following cross-section data were obtained: $\sigma (^7\text{Be(n,p)}_0) = 44900 \pm 1100 \text{(stat.)} \pm 3500 \text{(syst.)}$ b and $\sigma (^7\text{Be(n,p)}_1) = 43500 \pm 700 \text{(stat.)} \pm 1700 \text{(syst.)}$ b, respectively. Despite the slightly larger systematic uncertainties, these additional measurements verified the result discussed above. Combining all three data, a final value for the partial cross section of $^7$Be(n, p) reaction at thermal energy was found to be $43600 \pm 1600$ b.

To obtain the total cross section of the $^7$Be(n, p) reaction at the thermal energy, one should also add the contribution from the $^7$Be(n, p1) reaction. This reaction populates the first excited state of $^7$Li at 478 keV. The small peak at $\approx 750$ keV in Fig. 2 stems from protons from this reaction channel that have lost energy in the Al filter. The $\sigma_1/\sigma_0$ cross-section ratio is directly equal to the ratio of the $p_1$ and $p_0$ peak areas. The value of the $p_1/p_0$ ratio, obtained from the measurement with the 8.57 MBq $^7$Be sample, is 0.012(6). Although not very precise, this result is in accordance with the previous results by Bassi et al. [22], 0.02(1), Koehler et al. [6], 0.011(3) and Cervena et al. [5], 0.02(1).

Taking into account the above-mentioned $\sigma_1/\sigma_0$ ratio, the final result obtained in our experiment $44300 \pm 1400$ b is in accordance with the destruction cross section measured by Hanna [3] 48000 ± 9000 b, his (n, p) cross section evaluated via a gold standard and a $^{235}$U fission neutron flux monitor 53000 ± 8000 b, the (n, p) cross section values measured by Gledenov et al. [4] 50000 ± 10000 b, and by Cervena et al. [5] 46800 ± 4000 b. It should be, however, noted that all these previous results have significantly larger uncertainties. On the other hand, the result for $\sigma_0$ obtained in the current experiment 43800 ± 1400 b disagrees by more than three $\sigma$ with the value 38400 ± 800 b obtained by the Los Alamos Group [6]. The recent nTOF experiment that was optimized to cover a very wide neutron energy range rather than delivering a particularly precise value at thermal energies found a larger thermal cross section of 52300 ± 5200 b [7] that exceeds our value by 1.5 $\sigma$.

IV. SUMMARY AND CONCLUSIONS

Using a pure thermal neutron beam from a neutron guide, the thermal cross section of the $^7$Be(n, p0) reaction relative to the $^6$LiF standard was measured. Considering the 1/ν cross-section dependence for both ($^7$Be and $^6$Li) isotopes, the value $\sigma (^7\text{Be(n,p)}_0) = 43800 \pm 1400$ b for 25.3 meV neutrons was obtained. Because of rather low counting statistics, the ratio of the partial cross sections $\sigma_1/\sigma_0$ was obtained with a substantial uncertainty. It was found to be 0.012(6).

ACKNOWLEDGMENTS

The experiments were carried out at the “CANAM” infrastructure, as well as at the infrastructure of “Experimental nuclear reactors LVR-15 and LR-0” in Řež, both supported by the Ministry of Education, Youth and Sports under the Projects No. LM2015056 and No. LM2015074, respectively. The experiment was also supported by the Grant Agency of the Czech Republic under the Project No. P108-12G-108. We are grateful to the PSI crew A. Hagel, D. Viol, R. Erne, K. Geissmann, O. Morath, F. Heinrich, and B. Blau for the $^7$Be collection at the SINQ cooling system. This research was partially funded by the European Atomic Energy Community (Euratom) Seventh Framework Programme No. FP7/2007-2011 under Project CHANDA (Grant No. 605203). We acknowledge the support provided by the Federal Ministry of Education and Research (BMBF) through Grant No. 05K16PGA. We thank J. G. Correia (C2TN-IST, Lisbon University, Portugal) for help with the implantation at ISOLDE.