

Proceedings

# $n + {}^7\text{Be}$ Cross-Sections of Astrophysical Interest at the CERN n\_TOF Facility <sup>†</sup>

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† Presented at the 7th International Conference on New Frontiers in Physics (ICNFP 2018), Crete, Greece, 4–12 July 2018.

Published: 31 October 2019



**Abstract:** One of the possible explanations of the so-called Cosmological Lithium Problem (CLIP), i.e., the large overestimate of the primordial  ${}^7\text{Li}$  abundance by the standard Big Bang Nucleosynthesis theory (BBN), is related to an incorrect underestimation of the  ${}^7\text{Be}$  destruction rate by neutron-induced reactions. To verify this possibility, the  $n + {}^7\text{Be}$  reactions have been investigated at n\_TOF (CERN) in a wide neutron energy range, taking advantage of the new high-flux experimental area (EAR2) and specifically developed experimental setups. The  ${}^7\text{Be}(n, \alpha){}^4\text{He}$  cross section, measured for the first time from thermal to 10 keV neutron energy, was found consistent with previous estimates. A 10% increase of the  ${}^7\text{Be}$  destruction rate was instead determined on the basis of the  ${}^7\text{Be}(n, p){}^7\text{Li}$  cross section measured at n\_TOF from thermal to 300 keV neutron energy, a value clearly insufficient to provide a solution to the Cosmological Lithium Problem. Combined together, the two measurements finally rule out neutron-induced reactions on  ${}^7\text{Be}$  as a possible explanation of the CLIP.

**Keywords:** Cosmological Lithium Problem; Beryllium Abundance; Big Bang Nucleosynthesis

## 1. Introduction

Based on the Standard Model of electroweak and strong interactions, the Big Bang Nucleosynthesis (BBN) theory predicts the production of the lightest nuclides such as D,  ${}^3\text{He}$ ,  ${}^4\text{He}$ , and  ${}^7\text{Li}$  in the early Universe. While BBN calculations reproduce remarkably well, the primordial abundances of the lightest elements inferred from observations, a mismatch of a factor of three exists between predictions and the  ${}^7\text{Li}$  abundance observed in the photospheres of primitive, low-metallicity stars. This discrepancy, known as a Cosmological Lithium Problem (CLIP) [1], is one of the long-standing unresolved issues in Nuclear Astrophysics and Cosmology.

Several solutions have been proposed for the CLIP, some of which involve new physics beyond the Standard Model. From the point of view of Nuclear Physics, a large experimental effort related to Cosmology has been carried out in the past on a variety of subjects, including antiproton nuclear interaction in the early Universe, extensively studied by the LEAR and ASTROBELIX experiments [2–5]. A possible Nuclear Physics solution is related to an incorrect estimation of the destruction rate of  ${}^7\text{Be}$ . Since 95% of primordial  ${}^7\text{Li}$  is produced by electron-capture decay of  ${}^7\text{Be}$  (half-life  $t_{1/2} \simeq 53$

days), a higher rate for neutron or charged particle induced reactions on  $^7\text{Be}$  would lead to a lower  $^7\text{Li}$  abundance.

While charged particle induced reactions on  $^7\text{Be}$  have been accurately measured and the corresponding destruction rate calculated, the status of the experimental data on neutron induced reactions, i.e.,  $^7\text{Be}(n, \alpha)^4\text{He}$  and  $^7\text{Be}(n, p)^7\text{Li}$ , is quite unsatisfactory. This is mostly due to the very high specific activity of  $^7\text{Be}$  ( $\simeq 13 \text{ GBq}/\mu\text{g}$ ), which makes it difficult to procure and handle  $^7\text{Be}$  samples of adequate mass and purity.

On the  $^7\text{Be}(n, \alpha)^4\text{He}$  reaction, although its contribution to the destruction of  $^7\text{Be}$  has always been considered negligible in BBN calculations, no experimental confirmations of this assumption were available up to recently. At thermal neutron energy, only one measurement performed at the ISPRA reactor was reported in literature [6], which was used by Wagoner et al. [7] to extrapolate the cross section in the BBN energies range. Other theoretical calculations in the keV energy region differ significantly from each other [8]. A similar situation was also present for the  $^7\text{Be}(n, p)^7\text{Li}$  reaction; such a reaction represents 97% of the neutron destruction channel of the  $^7\text{Be}$  isotope and, as a consequence, it plays a key role in the determination of primordial lithium abundance. Despite its importance, the only two data sets covering from thermal to 500 eV [9] and from thermal to 13 keV ranges [10] show a systematic difference of up to 30% between each other. Furthermore, the extrapolation to higher energy is inconsistent with the cross section inferred from the time-reversal reaction, the  $^7\text{Li}(p, n)^7\text{Be}$  [11].

All these inconsistencies prompted a dedicated experimental program at the n\_TOF facility at CERN, taking advantage of the high-luminosity of the neutron beam, spanning from thermal to some GeV [12], at the recently built second experimental area (EAR2) [13]. The measurements, performed with two different silicon detector systems, provided for the first time reliable nuclear data on the  $n+^7\text{Be}$  reactions in a wide neutron energy range, allowing to clarify the role of these processes in the context of the Cosmological Lithium Problem.

## 2. The n\_TOF Facility at CERN

At the n\_TOF facility, neutrons are produced by spallation process on a lead target using 20 GeV/c protons/bunch with 7 ns RMS pulse width from the Proton Synchrotron (PS). Emitted neutrons, about 300 per incident proton, are subsequently collimated and guided through two beam lines at the two different experimental areas, EAR1 and EAR2, located at 180 m and 20 m from the spallation target, respectively. A layer of water (borated or demineralized) surrounding the spallation target moderates the initially fast neutron spectrum, providing a white neutron energy distribution spanning from a few meV up to the GeV region. The PS repetition rate below 0.8 Hz avoids the overlap of consecutive neutron bunches.

Both  $^7\text{Be}(n, \alpha)^4\text{He}$  and  $^7\text{Be}(n, p)^7\text{Li}$  reaction cross-sections have been measured in the second experimental area, n\_TOF-EAR2, characterized by an extremely high instantaneous neutron flux of  $10^7$  neutrons/pulse, 40 times higher than the one in EAR1 (see Figure 1). These characteristics make EAR2 ideal for measurements on isotopes available in very small amounts and with short half-lives, as is indeed the case of  $^7\text{Be}$ . More details on the EAR2 neutron beam can be found in Refs. [12,13].

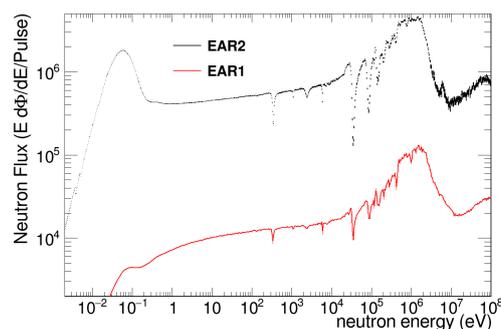


Figure 1. Comparison between the evaluated neutron flux in EAR2 (blue) and in EAR1 (red) [12].

### 3. The ${}^7\text{Be}(n, \alpha){}^4\text{He}$ Reaction

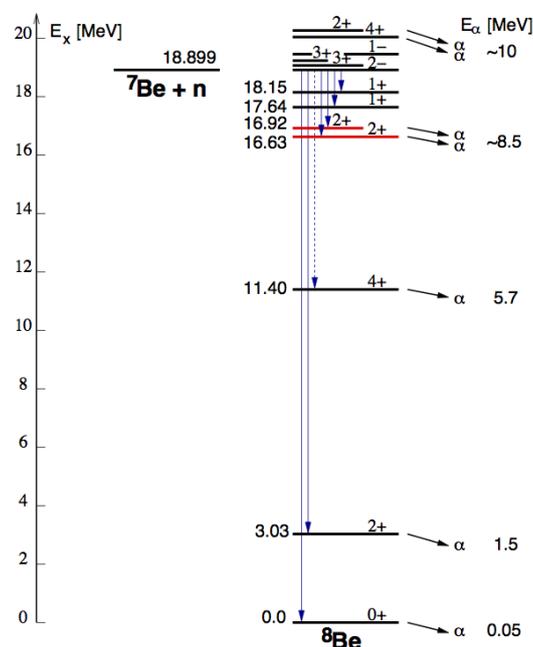
#### 3.1. The Experimental Setup

The samples for the  ${}^7\text{Be}(n, \alpha){}^4\text{He}$  measurement were prepared at the Paul Scherrer Institute (PSI), Villigen, Switzerland. Thanks to the high selectivity of the experimental setup to the two alpha particles emitted in the reaction, only a chemically pure sample material was required. Two samples of 3 cm in diameter were prepared with  $\simeq 18$  GBq of activity each; one was electro-deposited on a 5  $\mu\text{m}$  thick Al foil and the other one was droplet deposited on a 0.6  $\mu\text{m}$  thick low-density polyethylene foil. A more detailed description of the samples preparation is reported in Ref. [14].

The detection system consisted of two Si- ${}^7\text{Be}$ -Si sandwiches placed directly in the neutron beam. The Si detectors were  $3 \times 3 \text{ cm}^2$  in area and 140  $\mu\text{m}$  in thickness. The stack of detectors was placed in a 1 cm thick Pb shielded chamber for radio-protection purposes and the whole assembly was mounted in the vertical neutron beam of EAR2. Such a system allowed to detect in coincidence (within a 100 ns time window) the alpha particles emitted in the reaction, providing a peculiar signature of the occurrence of the  ${}^7\text{Be}(n, \alpha){}^4\text{He}$  reaction while rejecting all background components (see Section 3.2). Several performance tests and data quality checks have been performed prior to the measurement in order to verify its feasibility, the radiation resistance and background rejection capability of the detection system. The tests indicated an upper limit of 10 keV the neutron energy that could be reached in the measurement, due to the recovery time of the Si detectors after a “ $\gamma$ -flash” [13,15], the large flux of gamma-rays and relativistic charged-particles coming from the n\_TOF target.

#### 3.2. Data Analysis

The reaction proceeds through various energy levels of  ${}^8\text{Be}$  populated by gamma-ray transitions as shown in Figure 2. Taking into account that alpha particles in their ground state are bosons, for the total angular momentum and parity conservation rules allow only  $0^+, 2^+, 4^+ \dots$  states to decay.



**Figure 2.** Schematic level and decay scheme of  ${}^8\text{Be}$  transitions. The levels accessible experimentally in the n\_TOF measurement are indicated in red. Reprinted figure with permission from: Barbagallo et al. (n\_TOF Collaboration), Phys. Rev. Lett. 117, 152701—2016. Copyright (2016) by the American Physical Society.

Channels involving the ground state and the first excited level result in the emission of low-energy alpha particles ( $\lesssim 1.5$  MeV each), which could not be detected in the enormous 478 keV  ${}^7\text{Be}$  gamma-rays

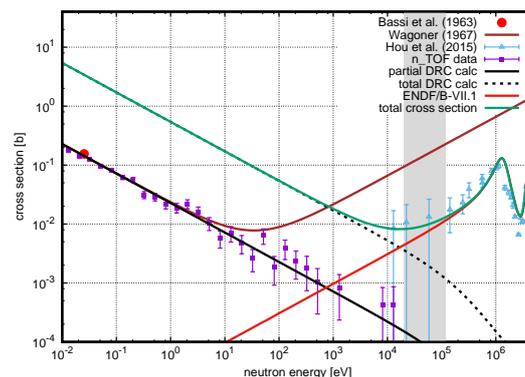
background. Moreover, the level  $4^+$  at 11.40 MeV can be populated only by second order magnetic transitions (M2) that are strongly suppressed. Therefore, only the partial cross sections involving the highest allowed excited levels ( $2^+$ ) between 16.6 and 18.1 MeV, which lead to the emission of two alpha particles with energy  $\gtrsim 8.5$  MeV each, are accessible experimentally.

The background component related to the gamma-rays from the  $^7\text{Be}$  decay corresponds to signals of a few tens of keV, although the large pile-up due to the very high activity could mimic a high-energy deposition event. A similar argument applies to the pileup of protons from the competing  $^7\text{Be}(n, p)^7\text{Li}$  reaction, characterized by a very large cross section at thermal neutron energy ( $\simeq 39$  kb). These background components were significantly reduced by applying a 2 MeV threshold on the signal amplitude.

The random coincidences produced by residual pileup events were estimated by analysing coincidence events of uncorrelated Si detectors (i.e., belonging to different arrays). Within statistical uncertainties, the count rates of both Si- $^7\text{Be}$ -Si arrays are constant in time, and on average two orders of magnitude higher than random coincidences during the whole measurement campaign.

### 3.3. Results

Figure 3 shows the measured partial cross section from 10 meV to 10 keV, obtained as a weighted average by uncertainties of the results from both arrays; data are affected by an overall systematic uncertainty lower than 10%, mainly due to the uncertainty of the sample mass and to the position of the neutron beam (see Refs. [15,16]). The result of this measurement at thermal energy was found to be consistent with the only previous existing measurement reported by Bassi et al.



**Figure 3.** The partial  $^7\text{Be}(n, \gamma)^4\text{He}$  cross section measured at n\_TOF (violet squares), compared with the corresponding one obtained by Direct Radiative Capture (DRC) calculations [17] (solid black curve) and the result of Bassi et al. [6] at 25.3 meV (red circle). The dashed black curve shows the calculated total cross section while the one suggested in Ref. [7] and used in BBN calculations is shown by the brown curve. A combination of the present results with evaluations of Evaluated Nuclear Data File, ENDF/B-VII.1 (red solid curve) provides the total cross section up to a few MeV (green curve). The hatched area shows the neutron energy region of interest for BBN. Reprinted figure with permission from: Barbagallo et al. (n\_TOF Collaboration), Phys. Rev. Lett. 117, 152701—2016. Copyright (2016) by the American Physical Society.

Theoretical calculations [17] were performed in order to estimate the branching ratios between different transitions, necessary to determine the total cross section from the partial one measured at n\_TOF. Since the n\_TOF data extend only up to approximately 20 keV, in order to determine the total cross section in the whole region of interest for BBN calculations, the n\_TOF data were combined with data from the indirect measurements of the  $^4\text{He}(\alpha, n)^7\text{Be}$  and  $^4\text{He}(\alpha, p)^7\text{Li}$  reactions [8], together with the evaluated cross section of ENDF/B-VII.1 [18] based on an R-matrix analysis of indirect reactions. The resulting cross section, from thermal neutron energy to a few MeV, is shown in Figure 3 (green

curve). In general, the new cross section shows a trend incompatible with Wagoner’s compilations used up to now for BBN calculations. In particular, the total cross section is more than a factor of ten higher at thermal neutron energy, but lower, by a similar factor, at high energy. The lower cross section in the energy range of relevance for BBN translates into a factor  $\simeq 10$  lower estimated rate for this reaction. As a consequence, these results indicate that the  ${}^7\text{Be}(n, \alpha){}^4\text{He}$  reaction cannot play any role in the Cosmological Lithium Problem, which in fact worsens by a few percent [16].

#### 4. The ${}^7\text{Be}(n, p){}^7\text{Li}$ Reaction

##### 4.1. The Experimental Setup

The measurement of the  ${}^7\text{Be}(n, p){}^7\text{Li}$  reaction required a completely different sample and detection concept. As for the  $(n, \alpha)$  reaction, the sample material was produced at PSI (Villigen, Switzerland) by radiochemical separation of 200 GBq  ${}^7\text{Be}$  from the cooling water of the Spallation Induced Neutron Source (SINQ) facility. After chemical purification, the material, in the form of nitrate solution, was inserted in the ion source of the ISOLDE facility at CERN, where an isotopically separated  ${}^7\text{Be}$  beam was produced and implanted on a 18  $\mu\text{m}$  aluminum backing. A sample of  $\simeq 1.1$  GBq total activity corresponding to  $\simeq 80$  ng in mass, with a purity of about 99% and 1% of  ${}^7\text{Li}$  contamination, was produced and immediately afterwards inserted in the EAR2 neutron beam line at n\_TOF (see Ref. [14] for more details). The  ${}^6\text{Li}(n, t){}^4\text{He}$  standard cross section [18] was used as a reference from thermal to 1 MeV neutron energy. To this purpose, a sample of 1.1 mg of LiF was prepared at INFN Laboratori Nazionali del Sud, Catania, Italy. To match the envisaged size of the  ${}^7\text{Be}$  sample, the lithium foil was masked with an aluminum opening window of  $1.5 \times 1.5$   $\text{cm}^2$  and used in the same experimental conditions of beryllium, as shown on the left side of Figure 4. Apart from being used as reference for the cross section measurement, the  ${}^6\text{Li}(n, t){}^4\text{He}$  reaction was used for calibrating the detectors, thanks to the known triton energy.

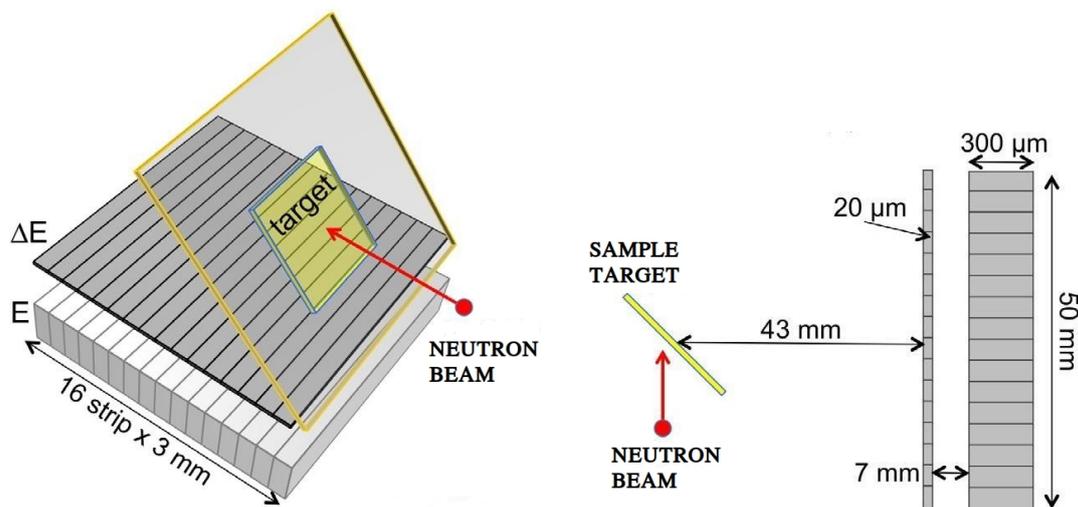


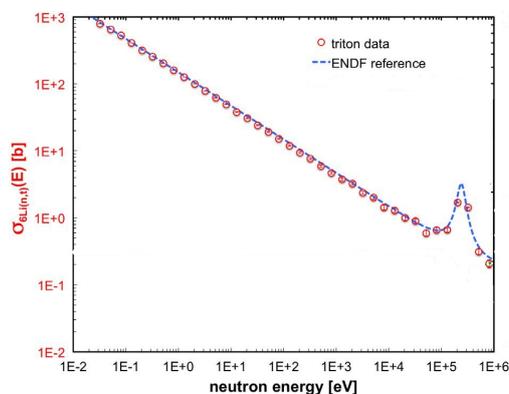
Figure 4. (right): 3D view of the detector setup; (left): profile view of the detector setup [19].

A position-sensitive telescope, made of two silicon strip detectors, of 20  $\mu\text{m}$  and 300  $\mu\text{m}$  thickness for  $\Delta E$  and  $E$  detection has been used (see right side of Figure 4); both silicon detectors had  $5 \times 5$   $\text{cm}^2$  active area and 16 strips. In order to minimize the energy straggling of emitted protons, the sample was tilted relative to the neutron beam direction by  $45^\circ$  (see right side of Figure 4). The whole setup was operated in a vacuum chamber, placed in the vertical beam line. Before the measurement, tests were performed to study the detector response and the expected background.

#### 4.2. Data Analysis

In order to identify protons emitted in the reaction, a coincidence between the strip in the  $\Delta E$  detector and the corresponding strips in the E detector was required. The width of the time coincidence window was chosen to be 100 ns. To extract the cross section, the geometrical efficiency of the telescope and the beam-sample convolution function needed to be determined. The detection efficiency of the setup was simulated by means of the Monte Carlo code GEANT4, in which a detailed software replica of the setup was implemented, including the samples and sample backing. The simulations were performed for different neutron beam energies, assuming a Gaussian shape reproducing the experimentally determined neutron beam profile. For both the cross section and angular distribution of emitted products, the GEANT4 High Precision model was adopted in the simulations, which uses the cross section and angular distribution of emitted products from evaluated data libraries. In the case of the tritons from the  ${}^6\text{Li}(n, t){}^4\text{He}$  reaction, the emission is isotropic up to a few keV, but becomes forward peaked at higher energies, leading to a drop in the detection efficiency. For the protons emitted in the  ${}^7\text{Be}(n, p){}^7\text{Li}$  reaction, evaluations assume isotropic emission at all neutron energies, resulting in a constant efficiency. Finally, simulations were also employed to calculate the beam-sample convolution factor, the sample dimension being smaller than the neutron beam profile, and in the case of the  ${}^7\text{Be}$  deposit, highly inhomogeneous.

The  ${}^7\text{Be}(n, p){}^7\text{Li}$  cross section was extracted relative to that of the  ${}^6\text{Li}(n, p)$  reaction, from the ratio of the number of normalized counts and taking into account the ratios of the efficiencies and beam-sample convolution factors. A check of the analysis procedure was performed by extracting from the n\_TOF data the  ${}^6\text{Li}(n, t){}^4\text{He}$  cross section. The very good agreement (better than 5%) found between data and the evaluations (see Figure 5) in the whole energy range validates the method used in the analysis and provides some indication on the related uncertainty.



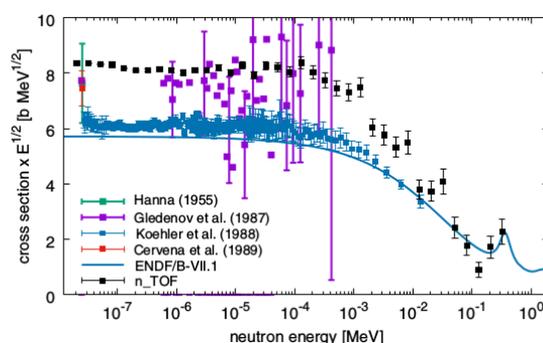
**Figure 5.** The  ${}^6\text{Li}(n, t){}^4\text{He}$  cross-section (circles) measured during the validation test, compared with the international standard (dashed line) [19].

One of the main background sources affecting the measurement is related to the  ${}^{14}\text{N}(n, p)$  reactions in the sample backing. In order to estimate this, a sample made of the aluminum backing has been used. This background was identified, subtracted, and its contribution was found to be important only for neutron energies above  $\simeq 330$  keV, a value representing the upper limit of the n\_TOF data. Other sources of systematics come from the detector efficiency that reaches almost 10% at 50 keV and 8% due to the inhomogeneity of the  ${}^7\text{Be}$  sample deposit. More details can be found in Ref. [14,20].

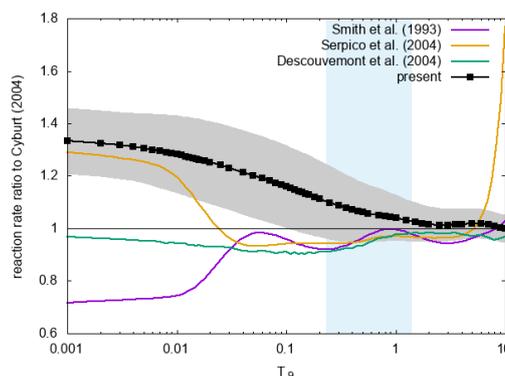
#### 4.3. Results

The  ${}^7\text{Be}(n, p){}^7\text{Li}$  cross section determined at n\_TOF is shown in Figure 6, compared with previous measurements and with the ENDF/B-VII.1 evaluated data. Present data are approximately 40% higher than those of Koehler [10] and ENDF evaluations, but at thermal energy they are consistent with the results of Refs. [9,21,22].

In order to extend the  ${}^7\text{Be}(n, p){}^7\text{Li}$  reaction cross section at BBN energies, data from Sekharan et al. [11] from 35 keV to 2 MeV on the time-reversal  ${}^7\text{Li}(p,n){}^7\text{Be}$  reaction have been used. The n\_TOF data, combined with the cross section extracted from the inverse reaction, have been fitted in Single Level Breit–Wigner approximation (SLBW) by means of an R-matrix code, in order to obtain a smooth cross section all the way from thermal neutron energy to a few MeV, covering in particular the energy region of interest for BBN calculations. Such cross section has then been convoluted with the Maxwellian neutron energy distribution corresponding to various stellar temperatures, and the resulting Maxwellian averaged cross section (MACS) has been used to calculate the astrophysical reaction rate, as a function of the temperature in units of  $T_9$  (i.e.,  $10^9$  K), in the range from  $10^{-3}$  to 10. In Figure 7, a comparison of the reaction rates of the present work with some of the previously adopted rates is shown. In all cases, the ratio of the calculated rate to a reference one reported by Cyburt et al. in Ref. [1] is plotted in the figure. The gray band corresponds to the experimental uncertainty of the reaction rate that was estimated to be around 10%. The vertical band represents the region of interest for BBN.



**Figure 6.** The cross section of  ${}^7\text{Be}(n, p){}^7\text{Li}$  compared with the previous measurements and ENDF evaluations. Only the statistical uncertainties are shown in the figure. Reprinted figure with permission from: Damone et al. (The n\_TOF Collaboration), Phys. Rev. Lett. 121, 04270—2018. Copyright (2018) by the American Physical Society.



**Figure 7.** Reaction rates of the  ${}^7\text{Be}(n, p){}^7\text{Li}$  normalized to that of Cyburt et al. [1]. The n\_TOF data are shown with the associated error band  $\simeq 10\%$ . Previously calculated rates are also shown in the figure. The temperature range of BBN with a larger impact on the lithium yield is indicated by the vertical band. Reprinted figure with permission from: Damone et al. (The n\_TOF Collaboration), Phys. Rev. Lett. 121, 042701—2018. Copyright (2018) by the American Physical Society.

Although the measured rate is significantly higher than previous evaluations in a wide range up to  $T_9 \simeq 1$ , it leads to at most a 12% decrease in the lithium abundance compared to previous calculations.

## 5. Conclusions

The Nuclear Physics solution for the CLIP sought in the  ${}^7\text{Be}(n, \alpha){}^4\text{He}$  and  ${}^7\text{Be}(n, p){}^7\text{Li}$  reactions has been investigated by measuring the two reactions in a wide energy range at the high-luminosity second experimental area (EAR2) of the n\_TOF facility at CERN. The  ${}^7\text{Be}(n, \alpha){}^4\text{He}$  results indicated that the corresponding reaction rate used in BBN calculation has been so far overestimated. On the contrary, the results on the  ${}^7\text{Be}(n, p){}^7\text{Li}$  reaction lead to a new estimate of the  ${}^7\text{Be}$  destruction rate 10% higher than currently believed, and thus in a corresponding decrease of the predicted lithium abundance. Both results therefore finally and unambiguously demonstrate that the  $n+{}^7\text{Be}$  reactions cannot solve the Cosmological Lithium Problem, ruling out one of the last possible Nuclear Physics solutions of this long-standing issue, while leaving open all other possible scenarios.

**Author Contributions:** Sipervisioning and responsible for the measurements, M.M. and M.B.; Formal analysis, M.B. and L.A.D.

**Funding:** 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).

**Conflicts of Interest:** The authors declare no conflict of interest.

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