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Cross-section measurement for the ^{7}Li(p,α)^{4}He reaction at proton energies 0.6 – 2 MeV

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ABSTRACT

Reliable data on the ^{7}Li(p,α)^{4}He reaction cross section are important for many applications, including fusion and accelerator neutron sources with a lithium target. Existing in the literature cross-section datasets in the literature are unfortunately inadequate and discrepant in many cases. In this study, the ^{7}Li(p,α)^{4}He reaction cross section is determined for proton energies E = 0.6–2 MeV. The experimental data are compared with the data from literature.

1. Introduction

The ^{7}Li(p,α)^{4}He reaction characterized by a high energy yield of 14.347 MeV is one of the thermonuclear reactions involved in the stellar cycle of fusion of heavy elements in the universe [1]. This reaction also accompanies the generation of neutrons in the ^{7}Li(p,n)^{7}Be reaction, which is used in a number of accelerator neutron sources [2] for boron neutron capture therapy of malignant tumors [3,4]. Knowledge about the ^{7}Li(p,α)^{4}He reaction cross section is certainly important for nuclear data evaluation. However, the existing cross-section datasets in the literature are unfortunately inadequate and discrepant in many cases [5–15]. Fig. 1 shows the data of ^{7}Li(p,α)^{4}He reaction cross section, and Fig. 2 shows the data of differential cross-section of this reaction.

The aim of this work is to measure cross-section of the ^{7}Li(p,α)^{4}He reaction.

2. Experimental facility

The study was carried out on an accelerator-based neutron source at the Budker Institute of Nuclear Physics in Novosibirsk, Russia [16]. The scheme of the experimental facility is shown in Fig. 3. The vacuum-insulated tandem accelerator *1* is used to provide a proton beam directed to a lithium target *6*. The proton beam energy can be varied within a range of 0.6 – 2 MeV, keeping a high-energy stability of 0.1%. The beam current can also be varied in a wide range (from 0.5 mA to 10 mA) with high current stability (0.4%). At the exit from the accelerator, the proton beam has a transverse dimension of 10 mm, an angular divergence of up to ± 1.5 mrad, and a normalized emittance of 0.2 mm mrad [17]. Proton beam current is measured and controlled by a non-destructive DC current transformer NPCT (Bergoz Instrumentation, France) *2*.

The proton beam was collimated to a ~ 10 mm diameter spot onto the target through a collimator of 1 mm in diameter *3*, placed at a distance of 4 m from the target, while the current did not exceed 1.5 μA on the target during all measurements.

The position and size of the spot onto the target were controlled by the Hikvision video camera *5* that detected the luminescence of lithium irradiated by protons [18]. The current of the proton beam hitting lithium was measured by a voltage divider using the target assembly *4* as a Faraday cup.

Lithium target was a thin layer of pure lithium deposited onto a copper substrate. Vacuum evaporation of lithium on the target was carried out at a separate stand. After lithium deposition, the target assembly *4* closed with a gate valve to maintain vacuum inside was disconnected from the lithium evaporation stand, transferred to the experimental facility and connected to the horizontal proton beam line.

The intensity and energy of α-particles in the ^{7}Li(p,α)^{4}He reaction were measured by the α-spectrometer *7* with silicon semiconductor

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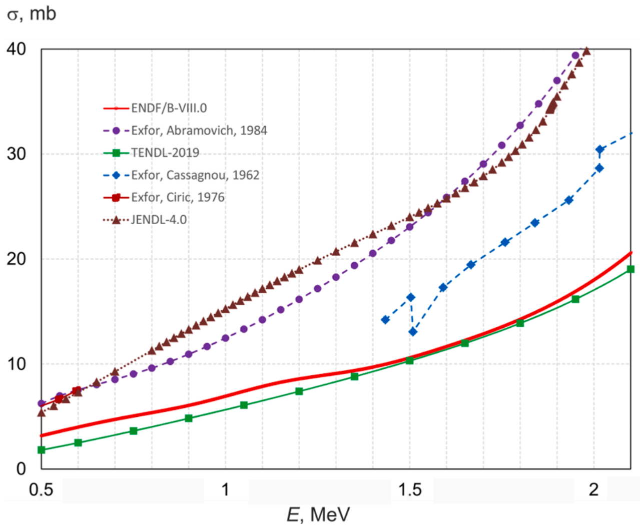

Fig. 1. The data of ^{7}Li(p,α)^{4}He reaction cross section presented in Java-based nuclear information software JANIS v.3.0 [5].

detector PDPA-1 K (Institute of Physical and Technical Problems, Dubna, Russia). Sensitive surface area of the detector was $S = 20$ mm^2, energy resolution – 13 keV, energy equivalent of noise – 7 keV, capacity – 30 pF, entrance window thickness – 0.08 µm, standard natural background in the range of 3–8 MeV – 0.15 imp/cm^{2}h.

When measuring the ^{7}Li(p,α)^{4}He reaction cross section, the sensitive part of the α -spectrometer detector was placed at a distance $R = 516$ mm from lithium at an angle of 168 ± 0.5° to the proton momentum. The solid angle was $\Omega_{lab} = S / R^2 = 7.51\cdot10^{-5}$.

3. Results and discussion

The ^{7}Li(p,α)^{4}He reaction cross section was measured as follows. A thin layer of lithium was irradiated with a proton beam and an α -spectrometer measures α -particles emitted at a certain solid angle. The differential cross section of the ^{7}Li(p,α)^{4}He reaction in the laboratory coordinates dσ/dΩ was found from the formula:

$$\frac{d\sigma}{d\Omega} = \frac{eY}{2kn l \Phi\,\Omega_{lab}},$$

where e – electron charge, Y – experimental yield of α-particles (integrated peak counts), number 2 indicates that the product of this reaction are two α -particles, k – efficiency of registration of α -particles by the spectrometer, n – density of ^{7}Li nuclei, l – lithium thickness, Φ – proton fluence, Ω_{lab} – solid angle.

The relationship of the differential cross section in the center of mass system $d\sigma_{c.m.}/d\Omega_{c.m.}$ and in the laboratory coordinates dσ/dΩ is given by the formula [19]:

$$\frac{d\sigma_{c.m.}}{d\Omega_{c.m.}} = \frac{|1+\beta cos\theta|}{(1+\beta^2+2\beta cos\theta)^{\frac{3}{2}}}\frac{d\sigma}{d\Omega},$$

where $\beta = \sqrt{\frac{m_p \widetilde{M}}{M_B M}\bullet\frac{T_M}{T_M+Q}}$ and $T_M = E_p\frac{M}{(m_p+M)}$, M, $\widetilde{M}$ – masses of decay particles, in this case the mass of an α -particle, m_p–proton mass, M_B – mass of the target particle, in this case mass of lithium, θ–particle detection angle in the laboratory coordinates, in this case 168°, Q–reaction energy yield, E_p – kinetic energy of incident proton. For convenience we introduce the coefficient connecting the coordinate systems G:

$$G = \frac{|1+\beta cos\theta|}{(1+\beta^2+2\beta cos\theta)^{\frac{3}{2}}}.$$

Since the radiation is isotropic in the center mass system, then the cross section of the ^{7}Li(p,α)^{4}He reaction σ was defined as:

$$\sigma = 4\pi G d\sigma/d\Omega.$$

It can be seen that to measure the cross section, it is necessary to know the efficiency of detection of α -particles by the spectrometer k, the density of ^{7}Li nuclei in the lithium layer n and the thickness of the lithium layer l, the solid angle Ω_{lab} in the laboratory coordinates, the coefficient connecting the coordinate systems G, as well as the proton fluence Φ.

Let us determine the detection efficiency k. The α -spectrometer was calibrated with two standard radiation sources based on the plutonium-239 radionuclide with activities of 4.01·10^5 Bq (passport No. 6887, marking 7165, 2P9-405.85, issue date 12.09.1985) and 1.21·10^5 Bq (passport No. 6882, marking 7160 1P9-105.85, issue date 12.09.1985). The confidence limits of the total error of the result of measuring the activity of each of their sources are set in the passports equal to 19 %. To determine the detection efficiency of the α -spectrometer, reference

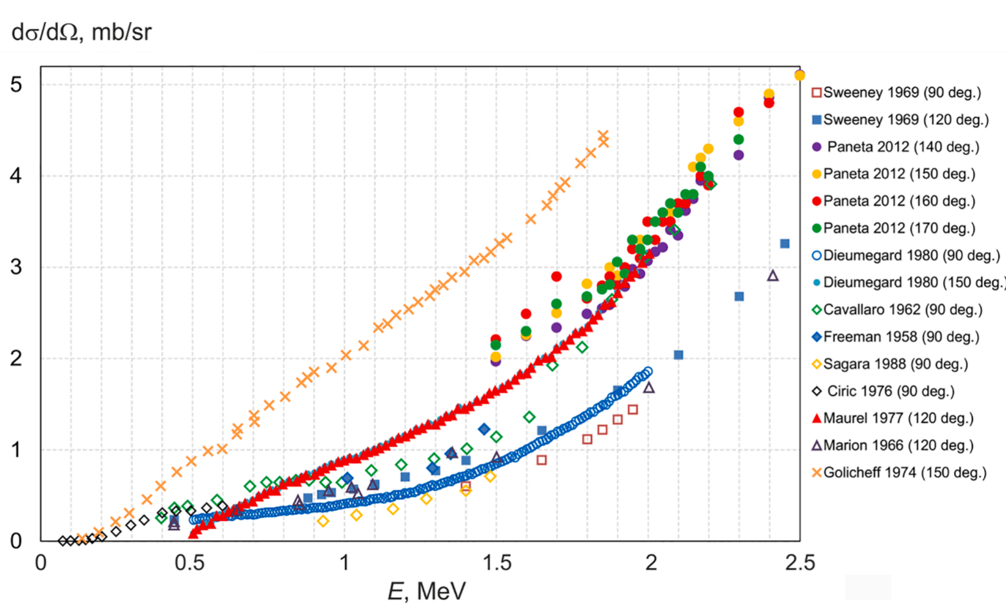

Fig. 2. The differential cross-section measurements for the ^{7}Li(p,α)^{4}He reaction [6–15].

radiation sources were placed at a distance of 102.8 mm from the detector surface and their activity was measured. The measured activities of the sources amounted to 4.16·10^5 Bq and 1.22·10^5 Bq – 4 % and 1.5% higher than the passport ones; they corresponded to passport values within the margin of error. Since the measured values of the source activity correspond to the passport ones, we considered the detection efficiency of α -particles by the detector to be 100 %, i.e. $k = 1$ with an accuracy of 3%.

The energy calibration of the spectrometer was carried out with an exemplary spectrometric α -source with the ^{226}Ra isotope (passport No. 425/331/10692-A, 21.10.1977) with an activity of 3.84·10^4 Bq, characterized by the energies of α -particles of the main transitions 4748, 5453, 5966 and 7651 keV. It was established that the dependence of the energy E on the channel number N was linear and was described by the expression E [keV] = 1.3578·N + 65.321.

Let us determine the density of lithium-7 nuclei in the lithium layer n. Previously, we found that the density of the evaporated lithium layer corresponded to the density of metallic lithium, equal to 0.54 g/cm^3 [20]. Taking the molar mass of lithium 6.997 g/mol and the density of lithium 0.54 g/cm^3, we got the volume of one mole of lithium equal to 13.1 cm^3. Dividing the Avogadro constant (6.022·10^{23} mol^{-1}) by this volume of one mole of substance, we obtained the density of lithium atomic nuclei equal to 4.596·10^{22} cm^{-3}. When lithium was evaporated, we used natural lithium produced by the Novosibirsk Chemical Concentrates Plant (Novosibirsk, Russia); in the batch used, the percentage of lithium was 99.956%. The percentage of lithium-7 in natural lithium varies from 92.41% [21] to 92.58% [22]; we assume the lithium-7 content equal to the average value, namely 92.50 ± 0.09 %. Consequently, the density of lithium-7 atomic nuclei in the lithium layer is $n = 4.596\cdot10^{22} \times 0.925 = 4.251\cdot10^{22}$ cm^{-3} with an accuracy of 0.1 %.

Let us determine the thickness of the lithium layer l. The thickness of lithium was measured by new *in situ* method [23]. The method is based on comparing the yield of 478 keV photons in the ^{7}Li(p,p′γ)^{7}Li reaction from the investigated lithium layer and from the thick one irradiated by 1.85 MeV protons.

A thick layer is a layer of lithium with a thickness greater than the length of the proton path in lithium up to the ^{7}Li(p,p′γ)^{7}Li threshold reaction equal to 0.478 MeV. When selecting the lithium thickness, we used the expression for the proton energy loss rate S in lithium depending on its energy E [24]:

$$S = \frac{S_{low} \bullet S_{high}}{S_{low} + S_{high}} \mathrm{eV}/(10^{15}\ \mathrm{atoms/cm^2}),$$

where $S_{low} = 1{,}6\,E^{0{,}45}$, $S_{high} = \frac{725{,}6}{E}\ln(1 + \frac{3013}{E} + 0{,}04578\,E)$, E in keV. By means of this formula, we obtain that the proton projected range in lithium is 145 μm for 1.85 MeV proton and 17 μm for 0.478 MeV (note that proton total path length and the projected range practically do not differ). Consequently, at the initial proton energy of 1.85 MeV, photons are generated up to a depth of 128 μm from the lithium surface.

A 227 μm thick lithium layer was evaporated on the target, for which 640 mg of lithium were used (for weighing, an analytical balance OHAUS PX-84 (USA) was used, the minimum weighing limit – 200 mg, the accuracy – 0.1 mg). The target was irradiated with a proton beam and the γ-ray spectrum was measured with a high purity germanium γ-ray spectrometer (SEG-1KP-IPTP 12 from Institute of Physical and Technical Problems, Dubna, Russia). The spectrum of γ-rays measured during 611 s (live time 600.3 s) at a proton beam current of 4.24 ± 0.02 μA is shown in Fig. 4a. The count rate of the 478 keV line is 84.59 count/s; per unit of current – 19.95 count/(s μA); integrated peak counts – 50757.

Then the lithium layer was washed off the target with water and a thin layer of lithium (approximately 0.5 μm thick) was deposited onto the target for which ~ 1.5 mg of lithium were used. The target with a thin lithium layer was placed in the same position, and the γ -ray spectrum was measured with a γ -ray spectrometer during 100 min (live time 5926.8 s) at a proton beam current of 10.10 ± 0.05 μA (Fig. 4b). The count rate of the 478 keV line is 1.50 count/s; per unit of current – 0.148 count/(s μA); integrated peak counts – 8904.

The ratio of the emission intensity of 478 keV photons per unit current from the studied lithium layer and from the thick one is A = 0.148 / 19.95 = 7.45·10^{-3}. Using the formula given in [24], we obtain the thickness of the studied lithium layer l (μm) = 45.698·A^2 + 56.281·A = 0.422. Taking into account the stability of the proton current (1 %), the statistical error of the data set (0.4 % and 1.1%), and the reliability of fitting the Gaussian distribution into the 478 keV line (0.5 % and 2.5 %), we obtain an error in measuring the lithium thickness of 3 %.

Thus, the thickness of lithium is l = 0.422 ± 0.013 μm.

Let us pay attention to the fact established by the method of energy analysis of backscattered protons [25]: in the residual vacuum, the

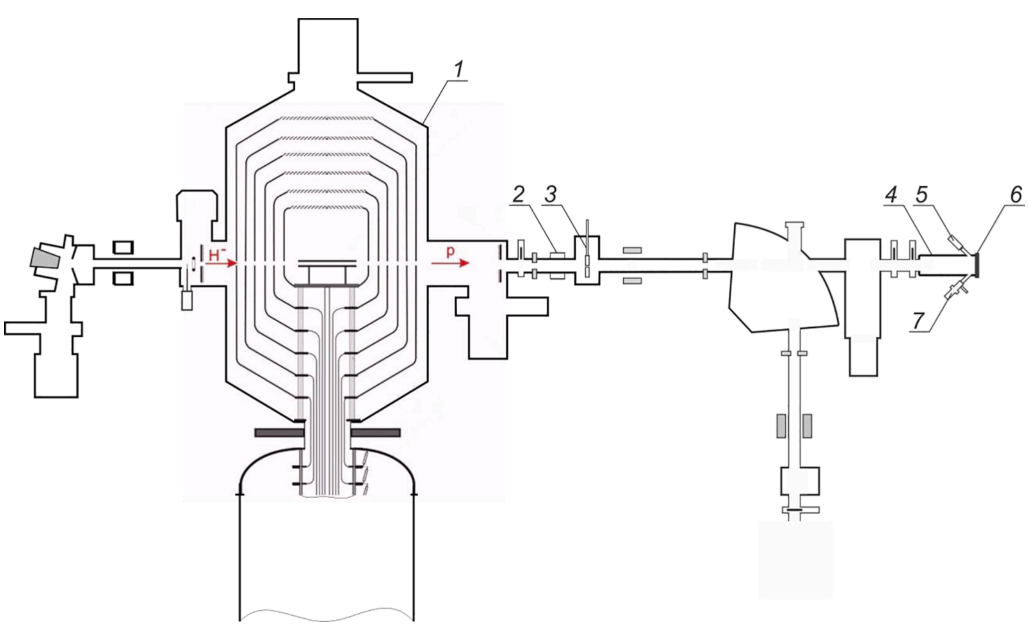

Fig. 3. Scheme of the experimental facility: *1* – vacuum insulated tandem accelerator, *2* – non-destructive DC current transformer, *3* – collimator, *4* – target assembly, *5* – video camera, 6 – lithium target, *7* – α-spectrometer.

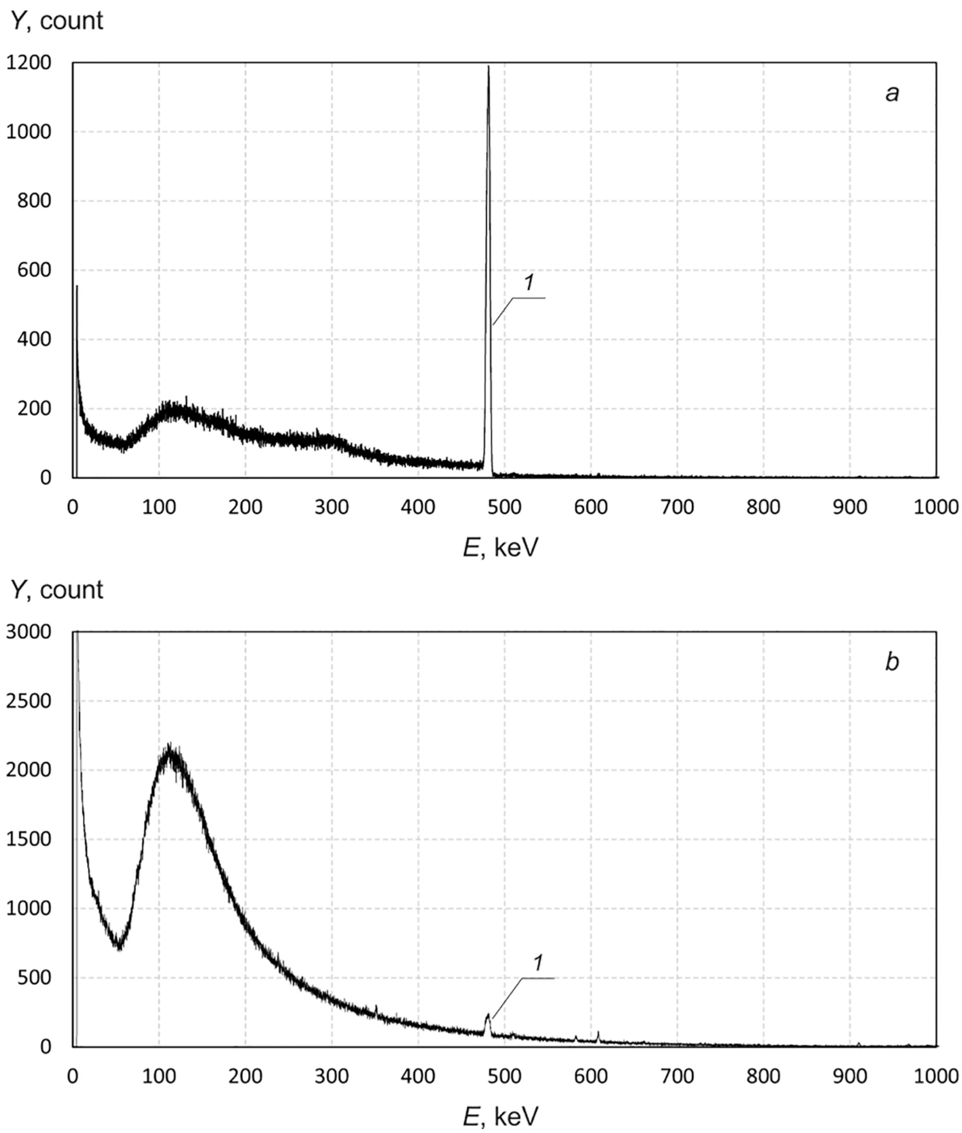

Fig. 4. The signal of HPGe γ -ray spectrometer: *a* – thick lithium layer, *b* – thin lithium layer.

lithium surface is covered with a thin layer of lithium oxide (several nanometers thick) and ~ 0.5 nm of carbon over it.

The characteristic spectrum detected by the α -spectrometer is shown in Fig. 5. The main signal (*1*, *2*, and *3* in Fig. 5a) is the signal of protons backscattered from copper atomic nuclei, where *1* are single events, *2* are double events, and *3* are triple events. Small peaks are clearly distinguished in the signal of single events, due to the scattering of protons on the atomic nuclei of lithium, carbon and oxygen (Li, C, and O in Fig. 5b).The spectrum of backscattered protons simulated by the SIMNRA v.7.03 program (Max Planck Institute for Plasma Physics, Germany) [26] with the above thicknesses of the lithium, carbon, and oxygen layers is in good agreement with the measured one. Signal *4* are the α -particles, signal *5* are the events of simultaneous registration of an α -particle and a backscattered proton.

The energy of α -particles depends on the energy of protons. Let's define this dependency. When a proton collides with an immobile lithium nucleus, a short-lived ($\tau \sim 10^{-18}$ s) compound nucleus will be formed, which will acquire momentum and, moving, decays into two α -particles. From the collision diagram, we determine the kinetic energy of the α -particle $E_M(\theta)$ detected at the angle θ in the laboratory coordinates [19]:

$$E_M(\theta)=E_p\frac{Mm_p}{(M+\widetilde{M})^2}\left[\cos\theta+\sqrt{\frac{\widetilde{M}(M+\widetilde{M})}{Mm_p}\left[\frac{M_B}{M_B+m_p}+\frac{Q}{E_p}\right]-\sin^2\theta}\right]^2,[0\leq\theta\leq\pi]$$

where *M*, $M^{\sim}$ – masses of decay particles, in this case the mass of an α -particle, m_p–proton mass, M_B – mass of the target particle, in this case mass of lithium, θ–particle detection angle in the laboratory coordinates, in this case 168°, E_p–kinetic energy of the incident proton, Q = 17.34 MeV–reaction energy yield. As the energy of the proton increases, the energy of the α -particle emitted at an angle of 168° decreases. So, if at a proton energy of 1 MeV the energy of an α -particle is 7.663 MeV, then at 2 MeV it is 7.523 MeV.

The maxima in the measured energy distributions of α -particles are obtained at energies lower than the calculated ones by 50–70 keV. Such a shift can be caused by ionization losses of the α -particle during the passage through the lithium layer. According to the Bethe-Bloch formula [27], the ionization loss of an α -particle in lithium is ≈ 600 MeV/(g cm^2) and passing through a layer of lithium with a thickness of 0.422 μm the α -particle loses an energy of ≈ 134 keV. Since α -particles are generated throughout the entire thickness of lithium they pass through lithium on average 0.211 μm and their average energy loss is 67 keV,

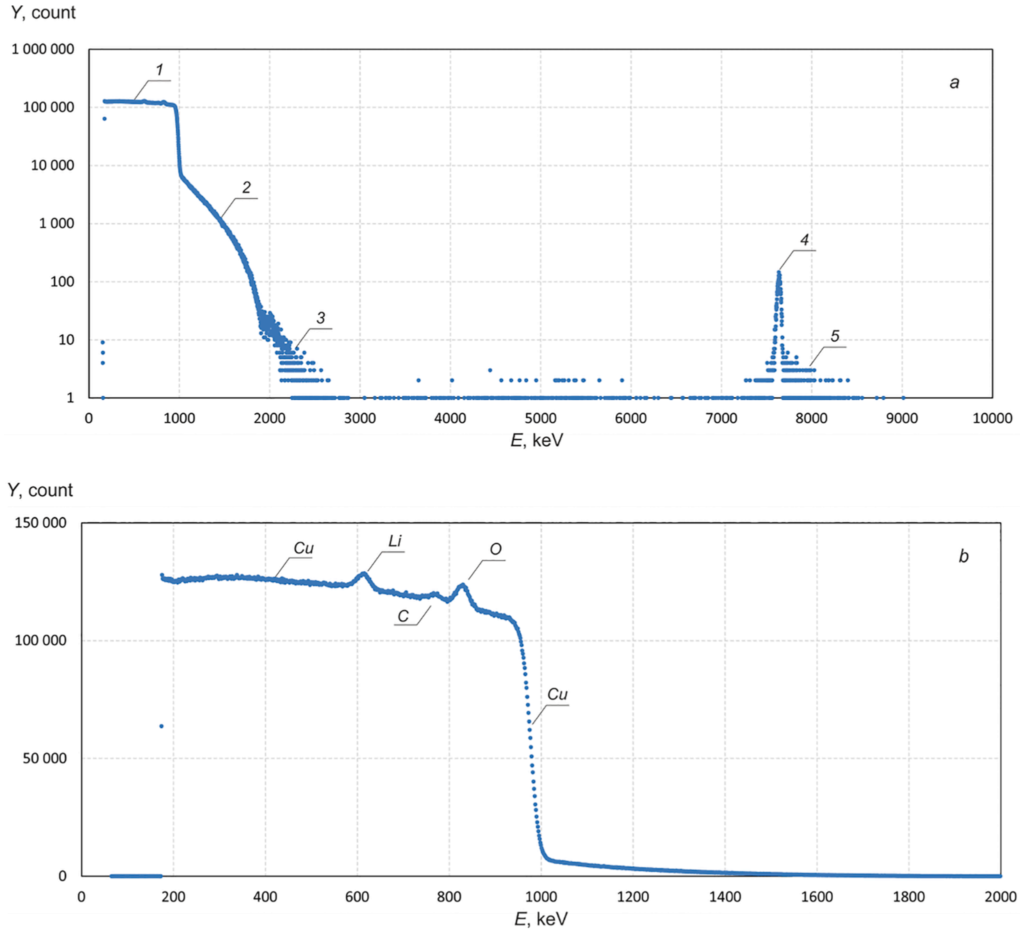

Fig. 5. The spectrum of charged particles detected by the α -spectrometer at a proton energy of 1 MeV: *1 – 3* – protons backscattered on copper atomic nuclei (*1* – single events, *2* – double, *3* – triple), *4* – α -particles, *5* – simultaneous registration of an α -particle and a proton. Cu, Li, C, and O are protons backscattered on the atomic nuclei of copper, lithium, carbon, and oxygen.

which is in good agreement with the measured shift in energy. Therefore, the measured shift in the energy of the α -particle is due to its ionization losses when passing through the lithium layer.

The measurements were carried out at 10 energy values. Table 1 shows the data on the measurements: the energy *E* and fluence Φ of protons, the total and live time of the measurement by the α -spectrometer, the experimental yield of α -particles (integrated peak counts) *Y*, the coefficient connecting the coordinate systems *G*.

To determine *Y*, all counts with particle energies from 6.5 MeV to 8 MeV were summed up and the natural background was subtracted. Since the standard natural background of the detector in the range of 3–8 MeV is 0.15 imp/cm^{2}h, and its area is 20 mm^2, then assuming a uniform distribution over the channels we obtain a background signal in the energy range from 6.5 MeV to 8 MeV equal to 0.9 imp/h. In 9 out of 10 experiments the measurements were carried out for one hour and it was believed that one background event was detected, which was subtracted. In one experiment the measurement was carried out for 0.24 h and it was considered that there were no background events during this time.

The obtained data on the cross sections are presented in Table 2 and Table 3. The results obtained are also shown in Fig. 6 for their comparison with the literature data given in Fig. 1 and Fig. 2. It can be seen that the available literature data on the differential cross section and the

Table 1
Data on the parameters of the measurements: *E* – proton energy, Φ – proton fluence, total and live times – total and live times of the measurement by the α -spectrometer, *Y* - experimental yield of α -particles (integrated peak counts), *G* - coefficient connecting the coordinate systems.

E, keV	Φ, mC	Total time, s	Live time, s	*Y*, count	*G*
601 ± 3	1.72	3756	3602	1300	1.2637
796 ± 4	3.79	3945	3613	4022	1.3115
900 ± 3	3.19	4008	3735	3838	1.3339
1000 ± 3	3.44	3908	3623	4536	1.3552
1104 ± 4	4.16	3987	3652	6186	1.3759
1300 ± 4	4.08	3916	3604	6939	1.4154
1503 ± 2	5.17	3981	3603	10,197	1.4531
1703 ± 4	1.55	3746	3634	3714	1.4977
1853 ± 2	0.96	873	817	2584	1.5149
2008 ± 15	4.20	4363	4075	13,363	1.5404

Table 2
The differential cross-section for the ^{7}Li(p,α)^{4}He reaction at 168°: *E* – the proton energy, Δ*E* – the standard deviation of *E*, σ – the cross section, Δσ – the statistical variance of σ.

E, keV	Δ*E*, keV	σ, mb/sr	Δσ, mb/sr
601	3	0.450	0.023
796	4	0.630	0.029
900	3	0.714	0.033
1000	3	0.784	0.035
1104	4	0.884	0.040
1300	4	1.010	0.046
1503	2	1.172	0.052
1703	4	1.425	0.066
1803	2	1.606	0.077
2008	15	1.892	0.084

Table 3
The ^{7}Li(p,α)α reaction cross section: *E* – the proton energy, Δ*E* – the standard deviation of *E*, σ – the cross section, Δσ – the statistical variance of σ.

E, keV	Δ*E*, keV	σ, mb	Δσ, mb
601	3	7.15	0.37
796	4	10.39	0.48
900	3	11.97	0.56
1000	3	13.35	0.60
1104	4	15.29	0.69
1300	4	17.96	0.81
1503	2	21.39	0.96
1703	4	26.82	1.25
1853	2	30.57	1.46
2008	15	36.62	1.63

total cross section differ significantly. Our differential cross section results are in agreement only with the data of Ciric [12], measured at an angle of 90°, and differ from all other data, including those measured at angles close to our 168°. At the same time, the data on the full cross section are in better agreement with the published data. In particular, they agree well with the Abramovich and JENDL data. It can be noted that our data on the cross section with good accuracy are exactly 2 times greater than the values given in the ENDF/B-VIII.0. We have no suggestion why the data of different authors differ so much.

4. Conclusion

Reliable data on the ^{7}Li(p,α)^{4}He reaction cross section are important for many applications, including fusion and accelerator neutron sources with a lithium target. Existing in the literature cross-section datasets in

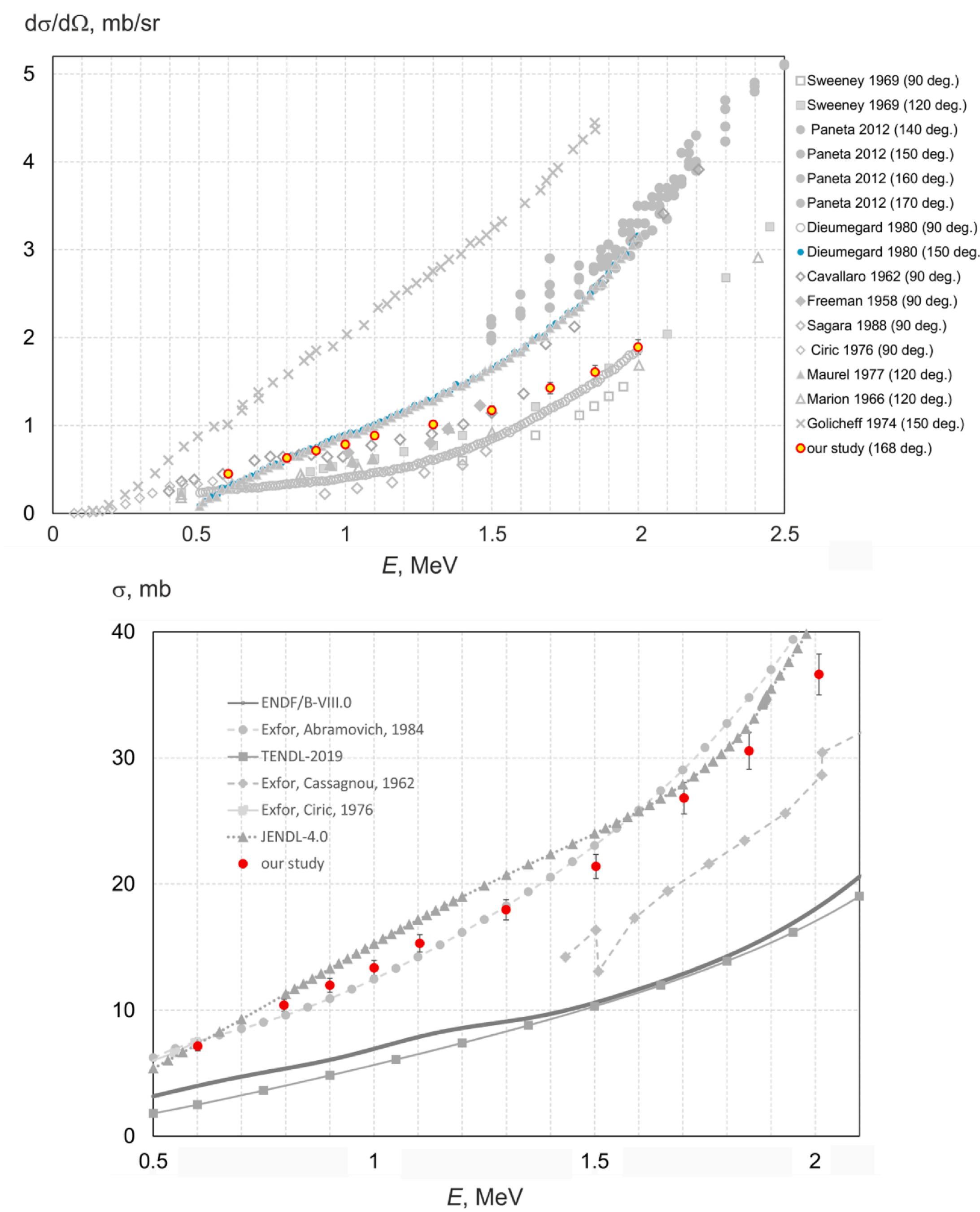

Fig. 6. The differential cross-section for the ^{7}Li(p,α)^{4}He reaction (*a*), the ^{7}Li(p,α)^{4}He reaction cross section (*b*).

the literature are unfortunately inadequate and discrepant in many cases. In this study, the $^{7}Li(p,\alpha)^{4}He$ reaction cross section at proton energies from 0.6 to 2 MeV was measured with high precision.

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CRediT authorship contribution statement

Sergey Taskaev: Conceptualization, Supervision, Writing – review & editing. **Marina Bikchurina:** Investigation, Writing – original draft. **Timofey Bykov:** Software. **Dmitrii Kasatov:** Investigation. **Iaroslav Kolesnikov:** Investigation. **Aleksandr Makarov:** Validation. **Georgii Ostreinov:** Investigation, Writing – original draft. **Sergey Savinov:** Validation, Investigation. **Evgeniia Sokolova:** Formal analysis.

Declaration of Competing Interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

Data availability

The theoretical and experimental data presented in this work are available from the corresponding authors on reasonable request.

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