

Contents lists available at ScienceDirect

Applied Radiation and Isotopes

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Revisiting the ⁷Li(p,n)⁷Be reaction near threshold

María S. Herrera^{a,b,c,*}, Gustavo A. Moreno^{d,e}, Andrés J. Kreiner^{a,b,c}

^a Comisión Nacional de Energía Atómica (CNEA), Av. Gral. Paz 1499, Buenos Aires B1650KNA, Argentina

^b Consejo Nacional de Investigaciones Científicas y Técnicas (CONICET), Av. Rivadavia 1917, Buenos Aires C1033AAJ, Argentina

^c Universidad Nacional de Gral. San Martín (UNSAM), 25 de Mayo y Francia, Buenos Aires B1650KNA, Argentina

^d YPF Tecnología S.A., Baradero, 1925, Ensenada, Buenos Aires, Argentina

^e Dpto. de Fl'isica, Facultad de Ciencias Exactas y Naturales, Universidad de Buenos Aires, Ciudad Universitaria, 1428, Buenos Aires, Argentina

HIGHLIGHTS

• We review neutron experimental data for the ⁷Li(p,n) reaction near threshold.

- A new computational method was used to study all the available published data.
- A consistent description of the neutron source was derived fitting the available data.
- We found that the neutron yield at 0° studied by Kononov is the most sensitive curve.
- A consistent set of parameters to parametrize the Breit-Wigner formula is presented.

ARTICLE INFO

Available online 27 November 2013

Keywords: ⁷Li(p,n)⁷Be reaction near threshold Differential neutron yield (p,n) Cross section

ABSTRACT

In this work we review all the available experimental neutron data for the ⁷Li(p,n) reaction near threshold which is necessary to obtain an accurate source model for Monte Carlo simulations in Boron Neutron Capture Therapy. Scattered published experimental results such as cross sections, differential neutron yields and total yields were collected and analyzed, exploring the sensitivity of the fitting parameters to the different possible variables and deriving a consistent working set of parameters to evaluate the neutron source near threshold.

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1. Introduction

The endothermic ⁷Li(p,n)⁷Be nuclear reaction near threshold can be used to produce low-energy neutrons with a forward-peaked distribution in the laboratory system which is suitable for Boron Neutron Capture Therapy (BNCT). The ground state Q-value of this reaction is – 1644.24 keV and the threshold value is given by a proton energy of 1880.29 keV yielding neutrons with 29.68 keV. This fact has motivated both theoretical and experimental studies of the ⁷Li(p,n) reaction near threshold (Zhou et al., 1997; Lee et al., 2000; Kenichi et al., 2002; Kononov et al., 2003; Bengua et al., 2006; Halfon et al., 2009; Kobayashi et al., 2011). However, most of the published cross section data for this reaction was measured several decades ago and only total neutron yield of the ⁷Li(p,n) reaction was measured recently with few data points below proton energies of 2 MeV.

In this work we review all the available experimental neutron data near threshold including differential cross section, differential neutron yield, total neutron yields and reported calculated data.

* Corresponding author. *E-mail address:* herrera@tandar.cnea.gov.ar (M.S. Herrera).

0969-8043/\$ - see front matter © 2013 Elsevier Ltd. All rights reserved. http://dx.doi.org/10.1016/j.apradiso.2013.11.042 These data sets are compared and analyzed searching for a consistent description of the neutron source.

2. Theory

The total cross section $\sigma_{(p,n)}$ near threshold in the center-ofmass (CM) system proceeding through a single resonance in the compound nucleus is given by the Breit–Wigner one-level formula (Gibbons and Newson, 1960)

$$\sigma_{(p,n)}(E_{p}^{CM}) = \pi \lambda_{p}^{2} g(J) \frac{\Gamma_{p} \Gamma_{n}}{(E_{p}^{CM} - E_{r})^{2} + \Gamma^{2}/4}$$
(1)

where Γ_p and Γ_n are the proton and neutron widths, respectively, and the total width is $\Gamma = \Gamma_p + \Gamma_n$. Here E_r is the resonance energy in the CM system.

When $(E_p^{\rm CM} - E_r)^2 \ll \Gamma^2$, it is possible to neglect the energy dependence in the denominator of Eq. (1) and the Breit–Wigner formula can be reduced to

$$\tau_{(\mathrm{p},\mathrm{n})} = 4\pi \lambda_{\mathrm{p}}^2 g(J) \frac{\Gamma_{\mathrm{n}}/\Gamma_{\mathrm{p}}}{\left(1 + \Gamma_{\mathrm{n}}/\Gamma_{\mathrm{p}}\right)^2} \tag{2}$$

where λ_p is the reduced de Broglie wavelength of the relative motion given by

$$\lambda_{\rm p}^2 = \frac{\hbar^2}{2\mu_{\rm i}E_{\rm p}^{\rm CM}} = \frac{\hbar^2(m_{\rm p} + m_{Li})^2}{2m_{\rm p}m_{\rm Li}^2} \frac{1}{E_{\rm p}}.$$
(3)

g(J) = (2J+1)/[(2S+1)(2I+1)] is the statistical factor that measures the probability that a particular compound nucleus state with angular momentum *J* will form according to the total spin of the projectile (*S*) and the target nucleus (*I*). Since for s-waves (*I*=0) the total angular momentum of the proton is only due to its intrinsic spin, one has $J^{\pi} = 2^{-}$, g(J) = 5/8 (Newson et al., 1957).

Particle channel widths are energy dependent and proportional to (Gibbons and Newson, 1960)

$$\Gamma \sim k v \gamma^2 \tag{4}$$

where *k* is the particle wave number, *v* its penetrability factor and γ^2 is the reduced level width. Thus, the ratio between the neutron and proton partial widths is

$$\frac{\Gamma_{\rm n}}{\Gamma_{\rm p}} \propto \frac{\sqrt{E_{\rm n}^{CM}}}{\sqrt{E_{\rm p}^{CM}}}.$$
(5)

For endoergic (p,n) reactions $\Gamma_n \propto (E_p^{CM} - Q)^{1/2}$, where Q is the absolute Q-value of the reaction. Transforming the above equation into the laboratory frame, and expression Q as a function of the proton energy at threshold (E_{th}), one obtains $x \equiv \Gamma_n / \Gamma_p \approx C \sqrt{1 - E_{\text{th}} / E_p}$, where C is a dimensionless parameter that has to be fitted from experimental data. Thus, evaluating the pre-factor in Eq. (2), the theoretical differential cross section can be described by

$$\frac{d\sigma}{d\Omega^{CM}}(E_{\rm p}) = \frac{169.72 \text{ MeV mb/sr}}{E_{\rm p}} \frac{x}{(1+x)^2},$$
(6)

where the proton energy in the laboratory frame (E_p) should be expressed in MeV. Notice that in this description *C* is the only parameter to be adjusted in order to match the data. This expression can only be used in the proton energy range of [E_{th} , 1.93] MeV, because above this region the CM differential cross section becomes directionally dependent and Legendre polynomials have to be included to account for the deviations. The coefficients have been reported only by Liskien and Paulsen (1975) and are important in the general case, however in this work we focus the discussion on the data near threshold.

In this paper we use a new algorithm based on the parametrization of the kinematical variables using center-of-mass and relative coordinates to compute both angular and energy distributions for thick targets. This new method has been validated using Lee and Zhou (1999a) and Kononov (2012) calculations, and is well suited to have an efficient access to all the variables in the problem allowing one to fit different observables easily. The program can also easily be recalibrated with new available experimental data.

The double differential neutron yield per proton current per time from a thick lithium target is related to the center-of-mass cross section via (Ritchie, 1976)

$$\frac{d^2 N}{dE_{\rm n} \, d\Omega} (E_{\rm n}, \theta) = \frac{f_{^7{\rm Li}} N_0}{eA_{eff}} \frac{1}{S(E_{\rm p})} \frac{d\sigma}{d\Omega^{\rm CM}} \frac{d\Omega^{\rm CM}}{d\Omega} \frac{\partial E_{\rm p}}{\partial E_{\rm n}}$$
(7)

where $f_{^7\text{Li}}$ is the ⁷Li atomic fraction of natural lithium (0.925), N_0 is Avogadro's number, e is the electronic charge, A_{eff} is the atomic weight of natural lithium and $S(E_p)$ is the proton mass stopping power in lithium (taken from SRIM, Ziegler, 1980). Here $d\Omega^{\text{CM}}/d\Omega$ and $\partial E_p/\partial E_n$ are the Jacobian transformations between the solid angles (CM and lab frame) and between proton and neutron energies (both in the lab system) respectively. Our method is based on the parametrization of all these quantities using CM and relative coordinates which gives an easy to use well defined numerical framework.

3. Neutron data available and discussion

In this section we review all the available experimental neutron data near threshold searching for a consistent calibration of the theoretical curve described in Section 2, Eq. (6). The (p,n) cross section, double differential neutron yield and total neutron yield data reported by different authors are analyzed.

The most recent measurement of the (p,n) cross section reaction was taken by Sekharan et al. (1976) for proton energies in the range [E_{th} , 4] MeV. However, the authors reported that older data near threshold published by Macklin and Gibbons (1958) and Gibbons and Macklin (1959) during 1958 and 1959, respectively, "is expected to be more nearly correct" in the older experiments due to a weaker dependence of the neutron energy detector efficiency near threshold. Therefore for the purpose of this work the Sekharan data will be disregarded.

Fig. 1 shows the cross section data measured by Newson et al. (1957) and by Macklin and Gibbons. The best fit least-squares of the analytical formula (Eq. (6)) to the data is obtained when $C=3.60\pm0.25$ (See Fig. 1). Lee and Zhou (1999b) applied the Breit-Wigner formula to Gibbons' data. However in their procedure the value *C* was fixed to C=6 and the pre-factor in Eq. (6) (which can be calculated a priori, as we did here) was adjusted. The author explains in his thesis (Lee, 1998) that this value was taken to be consistent with Newson's data but does not take into account an offset in the assumed threshold in Newson's work (Newson et al., 1957) (assumed threshold: 1881 keV, meanwhile Lee reported in his program 1880.25 keV), thus the resulted fit using the parameters reported by Lee (A = 164.913 MeV mb/sr and C=6) is acceptable in the flat region, but does not reproduce properly the initial rise of the data (see Fig. 1). Moreover, it should be pointed out that the best fit cannot capture the correct concavity in the reported data.

However, there exists another observable that can be used to corroborate the prediction of Eq. (6), and eventually fit *C*. In particular the double differential neutron yield at 0° measured by Kononov et al. (1977) is a good test for the analytical model. We have used our algorithm with a variable *C* value to fit this data. In this case two different data sets, S1 and S2, were generated. S1 corresponds to the entire experimental data points shown in Fig. 2, and S2 contains a subset of S1 without measurements in the neutron energy range



Fig. 1. Experimental data of the differential (p,n) cross section near threshold as a function of proton energy in the laboratory frame. The dashed line is calculated using parameters reported by Lee and the dotted line is the best fit to the data using Eq. (6).



Fig. 2. Double differential neutron yield at 0° as a function of neutron energy in the laboratory system (Kononov et al.). Solid lines are best fits to Eq. (6) using this data (S1 and S2). The differential neutron yield calculation reported by Lee (dashed line) and calculated using Newson's and Gibbons' data (dotted line) to determine *C* in Eq. (6) is also shown.



Fig. 3. Experimental values of total neutron yields for protons on natural lithium metal reported by different authors. Computed neutron yields using different sets of data for the theoretical $\sigma_{(p,n)}$ cross section are also shown.

[25.75, 44] keV to avoid possible deviations due to the sharp peak at threshold (measured values near threshold may deviate to lower values due to the proton energy spread). Using these sets the values obtained for C were $C_{S1} = 5.82$ and $C_{S2} = 6.00 \pm 0.05$ (assuming a Gaussian deviation for the set S2). Both estimations and, moreover, the curve computed with Lee's program represent fairly well the differential yield data of Kononov. However, it should be noted that the experimental data shown in Fig. 2 is not compatible with that shown in Fig. 1, because the best fit to Newson's and Gibbons' data generates a differential neutron yield at 0° in Fig. 2 that is incompatible with Kononov findings (and vice versa using the best fit with the Kononov's data, the cross section is almost the same as the curve shown in Fig. 1 with Lee's parameters). Judging both observables by the sensitivity in C and the consistent trend displayed by Kononov's data, we find the latter to be the most appropriate for a calibration of the analytical formula near threshold. This is also consistent with the fact that 0° is the angle for which the differential neutron yield has its maximum derivatives (sensitivity) with neutron energy. Total neutron yields have been measured by different authors in the range [*E*_{th}, 1.93] MeV (Lee et al., 2000; Lee and Zhou, 1999b; Yu et al., 1998; Kononov et al., 2006; Matysiak et al., 2011). This data is not so abundant and also more scattered than the previous observables. In particular, the experimental data published by Yu et al. will be disregarded because the values reported are two times lower than those obtained by other authors and no reasons causing this difference were reported (Yu et al., 1998). Fig. 3 shows the experimental data points available for proton beams on natural lithium in this range together with the different fits to the analytical cross section. Note that both ours and Lee calculations are compatible with the deviations reported in the experimental data, however, important differences between curves using Kononov's or Newson's and Gibbons' data are found, specially for the latter set as we move away from threshold.

4. Conclusion

In this work we have used a new computational method based on the center-of-mass and relative coordinate parametrization of the ⁷Li(p,n)⁷Be reaction to study all the available reported data near threshold. This method allowed us to provide new insight on the sensitivity of neutron production to the parameters in the model, giving an efficient tool to fit numerical predictions to published data. In particular we have shown that the differential yield of neutrons reported by Kononov et al. (1977) is the most sensitive curve when a Breit–Wigner formula is assumed to describe the threshold process. Moreover the best fit to this data was compared with the total neutron yields for protons on lithium showing a good agreement, thus providing a useful approach that can be used both to generate neutron sources near threshold and to incorporate new data points in future calibrations.

Acknowledgment

The authors wish to acknowledge Dr. O.E. Kononov for theoretical data used to validate the code and financial support from CONICET.

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