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Indications for a bound tetraneutron

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Using the reaction ^{7}Li(^{7}Li,^{10}C) we tried to populate states in the tetraneutron. A peak in the energy spectrum of identified ^{10}C, which we cannot attribute to a reaction with any other of the target components, corresponds to an excitation of the ^{10}C+4n system of 2.93 ± 0.16 MeV. Under different kinematic conditions an equivalent peak was observed. For a binding energy of the tetraneutron of -2.93 MeV a much larger width than the observed upper limit of $\Gamma < 0.24$ MeV (mainly due to experimental spread) is expected. Therefore, we favor the interpretation that this peak corresponds to ^{10}C in the first excited state at 3.354 MeV and a tetraneutron with a binding energy of $+0.42 \pm 0.16$ MeV.

1. Introduction

The nuclear force is the only force in nature where three-body effects have to be taken into account. Besides reactions between nucleons and deuterons, bound systems of few nucleons, like ^{3}H, ^{3}He or ^{4}He provide insight into three-body forces (e.g. [1]). But in these systems both kinds of nucleons are present. More desirable would be a system of three or more neutrons. Its binding energy and width would also constrain the properties of infinite neutron matter, i.e. of neutron stars. Already in the 1960s physicists were searching for a bound, or nearly bound, state of four neutrons, the tetraneutron (^{4}n) [2]. But no evidence for its existence was reported. Many different production methods have been studied (see also [3]), like double charge-exchange reactions on ^{4}He, e.g. (π^-, π^+), other reactions involving pions, three-proton pickup reactions on ^{7}Li, and production in fission of heavy nuclei. However, with the advent of short-lived radioactive beams indications for a ^{4}n state have been published. Marqués et al. [4] used the breakup of ^{14}Be beam particles in a carbon target and reported on six events consistent with a ^{4}n in the exit channel. To be detected, these should have lived for at least 60 ns to reach the 5 m distant detectors, and therefore practically be bound. However, a later analysis by some of the original authors allowed also a ^{4}n resonance up to a negative binding energy $-\mathrm{BE} < 2$ MeV [5]. More recently, Kisamori et al. [6] reported on a candidate resonant ^{4}n state produced in the double charge-exchange reaction ^{4}He(^{8}He,^{8}Be). They find that this state is unbound with respect to decay into four neutrons by 0.83 ± 1.41 MeV and has a width $\Gamma < 2.6$ MeV. Certainly, these claims are calling for independent experiments yielding more precise quantities.

2. Experimental details and results

Compared to secondary, radioactive beams, the use of stable beams has the advantage of many orders of magnitude higher intensity and much better defined momentum of the beam. The closest stable nucleus to ^{4}n is ^{7}Li and the removal of three protons is required. For the 3p-pickup an odd-even reaction partner will result in the least negative reaction Q-value. We chose the ^{7}Li(^{7}Li,^{10}C)^{4}n reaction because it has of all stable beams a moderately negative Q-value of -18.2 MeV to the just unbound four-neutron system. The concept of optimum Q-value (Q_{opt}) [7] is not applicable in our case, since it requires the ratio of Coulomb energy to kinetic energy not to change much in the moment of transfer, but the Coulomb energy is vanishing for the final channel. However, for ^{7}Li with our typical tandem energy of 46 MeV already the 2p-pickup has a large negative $Q_{opt} = -10.2$ MeV and a ground-state Q-value of -15.5 MeV, reasonably well matched. In addition, the ^{10}C ejectiles with about 23 MeV have nearly 70% probability to emerge from the target in the fully stripped charge

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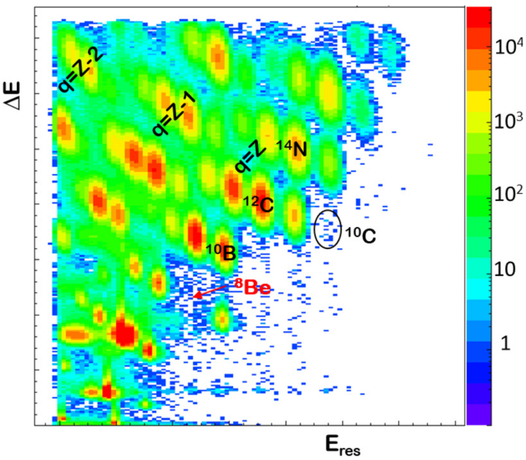

Fig. 1. Identification spectrum, energy loss vs. residual energy, for the run with a central ^{10}C energy of 20.5 MeV selected through the magnetic field. Clusters of ions with two, one and no electrons are denoted. For the completely stripped ions (q=Z) a few N=Z nuclei are labelled as well as the region of the ^{10}C ions.

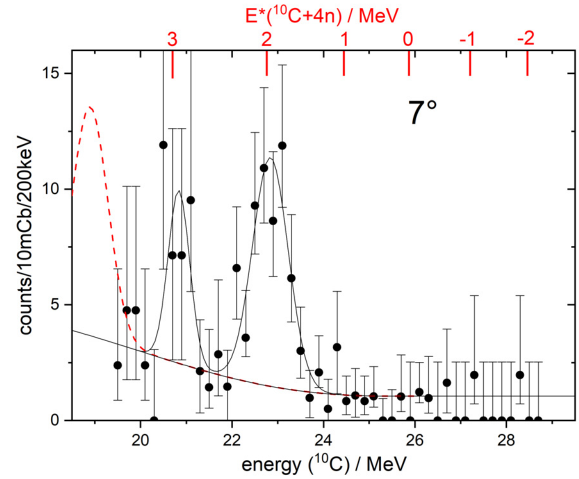

Fig. 2. Combined energy spectra of identified ^{10}C ions for six different magnetic field settings covering energies between 19.4 MeV and 28.4 MeV with the Q3D at 7°. The counts are normalized to the integrated beam current. On the top axis the scale for the total excitation energy of the ^{10}C+4 n system is shown (in red). Also shown is the fit with two peaks. In the fit a contribution from the phase space of four unbound neutrons in the exit channel and a constant background are included (solid line). The peaks fitted at 22.84(5) MeV and 20.84(10) MeV are interpreted as due to the ^{16}O(^{7}Li,^{10}C)^{13}B and the ^{7}Li(^{7}Li,^{10}C*)^{4}n reaction for a combined energy of ^{10}C excitation minus the binding energy of the tetraneutron of 2.93 MeV. The (red, dashed) peak at 18.9 MeV is drawn at the position where the ^{16}O(^{7}Li,^{10}C*)^{13}B reaction is expected.

state [8]. After we had prepared our experiment, we learned that a similar experiment had been published in 1974 [9] with non-conclusive result.

We produced ^{7}Li^{-} ions from fresh LiOH powder mixed with Nb in a Cs sputter ion source and accelerated them as ^{7}Li^{3+} to 46 MeV with the MP Tandem accelerator in Garching, near Munich. Typical beam currents were 50 pnA on the target. As targets we used nominally 100 μg/cm^2 thick layers of 99% enriched $^{7}Li_2O$ deposited on 20 μg/cm^2 C foils. Since the $^{7}Li_2O$ targets were quite hygroscopic after vapor deposition, there was a considerable amount of H_2O and possibly CO_2 bound in the target. On the other hand, in the course of irradiation there was also target material vanishing. This could be shown by an analysis using elastic recoil detection (ERD) with a ^{127}I beam [10] and by an energy loss measurement using an ^{241}Am α-source. Based on these measurements we used an effective thickness of 200 μg/cm^2 with equal atomic concentrations of Li, H and O to calculate the energy loss of the ions in the target. The ^{10}C ejectiles were momentum analyzed under a scattering angle of 7.0° in the Q3D magnetic spectrograph [11,12]. However, the Faraday-cup impeded forward angles up to 6° such that an angular range between 6.0° and 9.5° was accepted. The solid angle was about 9 msr. As detector in the focal plane we used a combination [13–15] of a single wire proportional counter for energy loss and position measurement and an array of (at the moment) 96 PIN Si-detectors, each 10 mm wide and 30 mm high for the residual energy and additional position measurement. Because of the large dispersion of the Q3D the roughly 1 m long detector covers an energy bin $\Delta E/E$ of only 10%. This results in a rather clean particle identification. The Si-detectors were read out individually, with the advantage that the amplification gain could be adjusted to correct for different response and dead layers. Because of the vanishing Coulomb repulsion in the outgoing channel the grazing angle in the lab system is only 1.2°, but at such small angles we could not use a Faraday-cup and the direct beam caused severe background. Still, we could expect some cross section at not so peripheral collisions. Fig. 1 shows an identification spectrum, energy loss (ΔE) vs. residual energy (E_{res}), for one setting of the magnetic field for ^{10}C with a central energy of 20.5 MeV. Due to the magnetic selection it shows ions in three charge states including the completely stripped ones at the highest E_{res}. The width in E_{res} is mainly caused by the angular acceptance. The locus of the $^{10}C^{6+}$ ions is indicated. The only condition required for this spectrum is the correlation between the position measured in the proportional counter and that in the Si-detectors. Under similar conditions we have recorded data for six different magnetic field settings and therefore kinetic energies and applied similar ΔE-E_{res} conditions as in Fig. 1. From the position measured with the proportional counter we have calculated the energy of the ^{10}C ejectiles. The fine binned (40 keV/bin) spectra are shown in the supplemental data [14]. In Fig. 2 the combined energy spectrum is drawn. Here we used the information from the number of the PIN diode and transformed it to a spectrum linear in energy. The average energy loss in the target (with an assumed thickness of 200 μg/cm^2 thickness) has been accounted for by using the program SRIM [16]. The number of events per 200 keV energy bin have been normalized to the same integrated beam intensity. The calibration of the magnetic rigidity of ions versus the position in the focal plane has been established by 46 MeV ^{7}Li^{3+} ions elastically scattered off a ^{12}C target at 7°. The asymmetric errors (due to the Poisson distribution) of the small numbers have been calculated according to the prescription of Feldman and Cousins [17] and we formed the weighted average of the data points in the overlapping regions. For the weight of the data points in the fits we used, in an iterative manner, the positive error bar if the fitted value was larger than the data point, and vice versa. We recognize only few events in the region above 25.88 MeV where the tetraneutron would be bound. But we find peaks at about 23 MeV and at about 21 MeV. For positive excitation energy E^* of the 4-n system we also have to consider the reaction leading to four unbound neutrons and the probability for that is governed by the phase space of the ^{10}C plus four neutrons which may be influenced by correlations, e.g. of pairs of neutrons. As already assumed by Kisamori et al. [6] for small excitations of the 4-n system and employed by Cerny et al. [9] we used an $(E^*)^3$ dependence. A larger exponent would increase the significance of the 21 MeV peak. A fit of such a phase space term, two Gaussians plus constant background to the whole spectrum, shown in Fig. 2, yielded peaks at 22.84(5) MeV and 20.84(10) MeV with height 9.9(1.2) and 7.5(3.0) and width of $\sigma_1 = 0.39(5)$ MeV and $\sigma_2 = 0.24(9)$ MeV, respectively (throughout we use 1σ uncertainties). The dominant uncertainty of the peak positions is due to the not well known energy loss of the ^{10}C in the target which we assumed as averaging 0.45 MeV at 21 MeV. We estimate a systematic uncertainty for the

Table 1
Q-values and kinetic energies of the ejectiles after (^{7}Li,^{10}C) reactions calculated for 46 MeV beam energy, different scattering angles and target nuclei; for the 4 n and 3 n systems binding energy $BE = 0$ is assumed, and for ^{4}n $BE = 0.42$ MeV. The 5th and 7th column are calculated for ^{10}C ejectiles in the first excited state at 3.354 MeV.

Target	Residue	Q (MeV)	7° E(^{10}C) (MeV)	7° E(^{10}C*) (MeV)	5° E(^{10}C) (MeV)	5° E(^{10}C*) (MeV)
^{7}Li	^{4}n	-17.75	26.43	20.81	27.13	21.88
^{7}Li	4 n	-18.17	25.88	19.70	26.59	20.96
^{6}Li	3 n	-10.92	33.43	30.06	34.16	30.88
^{12}C	^{9}Li	-25.75	17.26	–	17.57	–
^{16}O	^{13}B	-22.09	22.98	18.90	23.16	19.08
^{17}O	^{14}B	-25.27	19.28	14.70	19.44	14.89

peak positions from uncertain energy losses and a possible shift due to the kinematic correction of 0.4 MeV. The total spread of the energy loss of 0.9 MeV, corresponding to a variance of 0.26 MeV, also determines the width of the peaks. In order to see if these peaks can be produced by (^{7}Li,^{10}C) reactions on other target components, we look at Table 1. As was shown by the ERD analysis [14], the targets consisted only of the elements H, Li, C, and O. Compared with O, the N and F content was about 10^{-3}, that of B only 10^{-4}; the H of water vapor cannot contribute to the reaction. For the heavier isotopes ^{13}C and ^{18}O the (^{7}Li,^{10}C) reaction has a much more negative Q-value and ejectiles from the heavier isotopes will not interfere in the considered energy range. We expect the ejectiles from the ^{16}O(^{7}Li,^{10}C)^{13}B reaction at 23.0 MeV and can identify the 22.8 MeV peak with these. For this reaction the kinematic shift is over-corrected and could explain the larger width. The ^{17}O(^{7}Li,^{10}C)^{14}B would produce a peak at 19.3 MeV, but the abundance of ^{17}O is only 3.8×10^{-4}. The ^{10}C energy closest to 20.8 MeV from reactions on the significant target components would be 18.9 MeV (see Table 1) from the ^{16}O(^{7}Li,^{10}C*)^{13}B reaction. A peak at that energy with the same width and height as that from the ^{16}O(^{7}Li,^{10}C)^{13}B reaction is added (in red) in Fig. 2. Obviously, it would not influence the region of the 20.8 MeV peak.

Thus, we cannot attribute the peak at 20.8(4) MeV to ^{10}C ejectiles from any other target component than ^{7}Li. Therefore, we attribute this peak to the ^{7}Li(^{7}Li,^{10}C)^{4}n reaction. This ejectile energy corresponds to a total excitation energy of the ^{10}C + 4n system of 2.93(16) MeV. The height of the peak is 2.5 times its uncertainty. But there is a better way to estimate the significance of the 20.8 MeV peak. The data up to an energy of 21.2 MeV were obtained in a single setting of the spectrograph field. We make use of the Poisson distribution of the four bins 20.5 to 21.1 MeV which have a total of 15 events. The sum of the four values from the fit with two lines (not normalized to the integrated beam current) is 12.70. If we fit the spectrum with just one peak, the 22.8 MeV peak barely changes, but mainly the factor of the phase space contribution. Then, the sum of the four values at the postulated 20.8 MeV peak amounts to 5.51. The probability to observe 15 or more events from a Poisson distribution with expectation value 5.51 is $6.1 \cdot 10^{-4}$, while it is 0.295 for an expectation of 12.70. The likelihood ratio amounts to $2.1 \cdot 10^{-3}$ or the significance of the 20.8 MeV peak to roughly 3σ.

The width of the peak of $\sigma = 0.24(9)$ MeV can well be explained by the energy loss differences in the target, which amount to about 0.9 MeV, imperfect focussing and imperfect compensation of the kinematic shift. A spread and shift of the focal plane position is possible because the ejectiles of the ^{7}Li(^{7}Li,^{10}C)^{4}n reaction have a strong kinematic shift (change of kinetic energy as a function of scattering angle, see Fig. 3) and the dipole component of the magnetic multipole element between the first two dipoles [18] was adjusted to remove this shift (mostly the linear term and not perfectly - because of a lack of experience with such a strong kinematic shift). In Fig. 3 we have also calculated how the correction with the quadrupole field component of the multipole shifts the ^{10}C peaks for the ^{7}Li(^{7}Li,^{10}C)^{4}n reaction at $E^* = 2.93$ MeV what leads to an overcompensation for the ^{16}O(^{7}Li,^{10}C)^{13}B reaction.

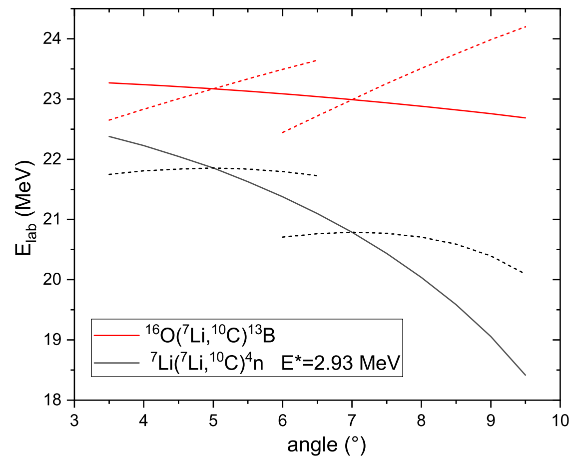

Fig. 3. Angle dependence of the two reactions ^{16}O(^{7}Li,^{10}C)^{13}B and ^{7}Li(^{7}Li,^{10}C)^{4}n for a total excitation energy of the ^{10}C plus 4 neutron system of 2.93 MeV. In the dashed curves the kinematic correction (only linear term) for the ^{7}Li+^{7}Li reaction and for the spectrograph at 7° and at 5° is taken into account. For the ^{16}O(^{7}Li,^{10}C)^{13}B reaction the shift is then overcompensated.

The width in the particle energy corresponds to an upper limit for the width in the ^{4}n binding energy of 0.10 MeV, or $FWHM = 0.24$ MeV. The intrinsic width of the state must be small compared to the latter value. An intrinsic width $\Gamma < 0.24$ MeV for a ^{4}n state unbound by 2.9 MeV seems unrealistic. Experimentally, we can compare it with the 4-particle decay (also with vanishing Coulomb barrier) of ^{6}H→^{3}H+3 n which has a Q-value of 2.72 MeV and a width of 1.55 MeV, at least six times larger. Theoretically, e.g. Shirokov et al. [19] calculated a ^{4}n resonance at an energy $E_r = 0.8$ MeV and a width $\Gamma = 1.4$ MeV; Li et al. [20] obtained $E_r = 2.64$ MeV and $\Gamma = 2.38$ MeV. Thus, we tend to believe that the ^{10}C ejectile is in the first excited state at $E^* = 3.354$ MeV and that the ^{4}n state is bound by 0.42(16) MeV. In that case the width of the ^{10}C peak would be broadened by the recoil from the emitted γ-ray [21]. But the maximum shift of ±0.22 MeV and its distribution, depending on the distribution of the populated m-substates, could still be enclosed in the observed width. The second excited state of ^{10}C at 5.2 MeV is no choice. It lies 1.2 MeV above the p-threshold and has a mean life of 3×10^{-21} s. The intensity of the 20.8 MeV peak corresponds to a cross section of 30 nb/sr. Although events from ^{10}C in the ground state and ^{4}n bound by 0.42 MeV should have 26.4 MeV, we consider the events above 25 MeV as background with an upper limit for a peak of 3 nb/sr. Due to the variation of the targets during irradiation, cross sections have an uncertainty of a factor of two.

A possible explanation for the preference of the excited ejectile is the small energy available in the center-of-mass (CM) system of 23 MeV leaving just 1.8 MeV kinetic energy for the ejectiles after the reaction that we postulate. Thus, there is barely energy left to excite and split the ^{4}n apart. Another reason is the multistep transfer which implies some sticking between the reaction partners and the creation of angular momentum. When irradiating a LiF target in the 7° geometry we observed the ^{19}F(^{7}Li,^{10}C)^{16}C* reaction at 28 MeV ^{10}C energy (see supplementary material [14]). Here ^{16}C is in the first excited (2^+) state at 1.77 MeV. Before the Garching accelerator lab was shut down in January 2020 we had a last chance to check our observation. There we used a smaller Faraday cup and could position the Q3D at 5° hoping that the cross section might increase and with the intention to modify the kinematics compared to the earlier experiment. The accepted an-

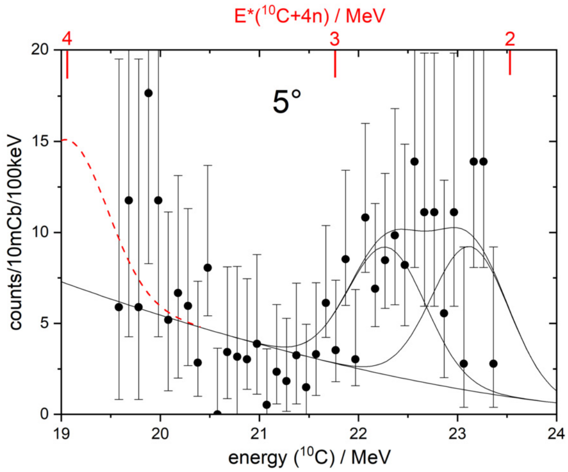

Fig. 4. Combined spectrum of ^{10}C nuclei detected at 5°. Each data point is the weighted average of up to four values for different settings of the magnetic field. Also drawn is a fit of the (^{10}C+4-n) phase space and two Gaussians with width 0.39 MeV at the positions 23.1 MeV and 22.3 MeV as well as their sum. These correspond to the same reactions as in the 7°data of Fig. 2. Again, the (red, dashed) peak at 19.1 Mev is drawn at the position where the^{16}O(^{7}Li,^{10}C*)^{13}B is expected. All data points are used for the fit.

gular range was ±1.5°, the solid angle 7.4 msr. The disadvantage of the more forward angle can be seen in Fig. 3: the ^{10}C energies for the ^{16}O(^{7}Li,^{10}C)^{13}B and the ^{7}Li(^{7}Li,^{10}C*)^{4}n reaction come much closer. The resulting spectrum from averaging the data for four overlapping magnetic field ranges is shown in Fig. 4. The structure above 21 MeV can be fitted by a single peak. But the resulting width of $\sigma = 0.71(10)$ MeV seems too broad, if compared with the widths observed in the 7° data and considering that the kinematic shift is smaller at the smaller angle. However, we could convince ourselves that the data sets for the two beam times are consistent by fitting the 4-n phase space and two Gaussians with common width (since the two peaks strongly overlap, the correlation between the parameters is too strong when we fit with independent widths) to the complete data set, varying again their energy, height, common width, but no background, since that is not defined on the high-energy side. The common width is fitted to 0.39(14) MeV, similar as for the 7° data set. One peak is fitted at 23.13(34) MeV - the uncertainty increases to 0.5 MeV, if we add in quadrature the 0.4 MeV systematic uncertainty - and corresponds exactly to the ^{16}O(^{7}Li,^{10}C)^{13}B reaction. The other peak is fitted to 22.29(22) MeV resulting in 22.3(5) MeV when including the systematic error. This is consistent with the energy 21.91 MeV expected for the ^{7}Li(^{7}Li,^{10}C*)^{4}n reaction with a total excitation energy of 2.93 MeV. Its height of 7.0(2.5) counts/10mCb/100 keV corresponds to a cross section of 60 nb/sr, perhaps consistent with a rising cross section towards 0°. The intensity of the phase space contribution is about 4 times larger than at 7°. Some of the count rate towards low energy could be due to the ^{16}O(^{7}Li,^{10}C*)^{13}B reaction, expected at 19.1 MeV, where we have drawn in Fig. 4 a peak with the same height as for the ^{16}O(^{7}Li,^{10}C$_{gs}$)^{13}B reaction. Peaks from the^{16}O(^{7}Li,^{10}C)^{13}B* reaction are expected a little lower in energy.

3. Comparison with earlier experiments and theory

The same ^{7}Li(^{7}Li,^{10}C)^{4}n reaction has been explored at higher beam energy, 79.6 MeV, in the 1970ies by Cerny et al. [9]. They used similar targets as we and detected the ejectiles at 7.4°. Later on, the ^{7}Li(^{7}Li,^{10}C)^{4}n reaction has been examined [22] under nearly the same conditions: beam energy 82 MeV and detection angle 2°. Neither of these experiments did observe an indication of a ^{4}n state. In the Cerny et al. spectrum [9] the peak corresponding to a total excitation energy of 2.93 MeV would come at a ^{10}C energy of 56.2 MeV, too close to the ^{16}O(^{7}Li,^{10}C)^{13}B peak at 56.8 MeV. But Aleksandrov et al. [22] seemed to have avoided the ^{16}O in the target and in their spectrum there is only a small peak visible at the position marked by "^{13}B". We calculate the ^{7}Li(^{7}Li,^{10}C*)^{4}n peak for a total excitation of 2.93 MeV only 0.7 MeV higher and thus barely distinguishable. They quote an upper limit for the bound ^{4}n cross section of 0.1 nb/sr in the CM system, but none for finite excitation. Our lab cross sections are enhanced by a factor of about 50 relative to the CM values; the CM values of 0.6 and 1.2 nb/sr are considerably larger than the upper limit of Aleksandrov et al. for bound ^{4}n, but it is not clear, how much for 3 MeV of excitation. However, as mentioned above, the main difference between ours and the other two ^{7}Li+^{7}Li experiments is the energy available in the CM system. With a CM energy of 23 MeV we are just 4.8 MeV above the threshold for ^{10}C + 4 neutrons and just 1.8 MeV above ^{10}C* plus the bound ^{4}n that we postulate. Aleksandrov et al. are 20 MeV above that energy and the much larger available energy could be the reason why in their case the brittle ^{4}n does not survive; and it may help that the reaction partner in our case takes up most of the available energy.

Theoretically, there have also been many efforts to calculate the energy of a 4-neutron resonance, especially in recent years triggered by the Kisamori et al. observation [6]. Ab-initio no-core shell-model calculations have been used [19,20,23] as well as chiral effective field theory [24] and continuum calculations [25,26]. But most of them [23,25,26] come to the conclusion that such a system should be unbound by a few MeV and accordingly have a large width. Lazauscas et al. [27] have tried to reproduce the double charge-exchange reaction [6] leading to a ^{4}n resonance, with no success. However Shirokov et al. [19] get a ^{4}n resonance at an energy $E_r = 0.8$ MeV, Gandolfi et al. [24] with $E_r = 2.1$ MeV and Li et al. [20] at nearly $E_r = 3$ MeV. The continuum calculations [25,26], although denying a resonance, find a low-energy enhancement of the density of states which might explain the Kisamori et al. [6] observation. Already earlier, Pieper et al. [28], using a Greens-function Monte-Carlo approach, had stated that our understanding of nuclear forces would have to be significantly changed to accommodate a bound or nearly bound ^{4}n. But, perhaps, that may be necessary.

4. Conclusions

In conclusion, we have observed a peak in the ^{7}Li(^{7}Li,^{10}C)4n reaction in the angular range 6°-9.5° which corresponds to an excitation energy in the final channel of 2.93(16) MeV. A measurement at 5° showed an equivalent peak at the same excitation energy of the ^{10}C+4n system, although overlapping with that from the ^{16}O(^{7}Li,^{10}C)^{13}B reaction. This leaves the two possibilities: a ^{4}n state unbound by 2.93 MeV and an extraordinarily small width $\Gamma < 0.24$ MeV or the ^{10}C is in the first excited state and the ^{4}n has a bound state with a binding energy of BE=+0.42(16) MeV. The latter option supports the claims of ref. [4,6]: the positive binding energy favors a bound ^{4}n as suggested by Marqués et al., [4] and our binding energy agrees with the value of Kisamori et al. [6], but is nine times more precise. If the binding energy of the trineutron is smaller (and that of the dineutron smaller than half of it) this state can only decay by β-decay to ^{4}H which subsequently would emit a neutron. The β-decay Q-value would be 7.27 ± 0.19 MeV. Since, as every even-even nucleus, ^{4}n has the ground-state spin/parity 0^+ and that of ^{4}H is 2^-, the β-decay would be first forbidden unique. More favorable would be the non-unique first forbidden decay to the first excited 1^- state at 310 keV. Using a typical $\log ft$=7.0 [29] we estimate [30] for ^{4}n a half-life around 450 s, comparable to that of the neutron.

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.

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Appendix A. Supplementary material

Supplementary material related to this article can be found online at https://doi.org/10.1016/j.physletb.2021.136799.

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