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$^{17}O(n,\alpha)^{14}C$ cross section from 25 meV to approximately 1 MeV

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We have measured the $^{17}O(n,\alpha)^{14}C$ cross section from thermal energy to approximately 1 MeV. A bump in the data near 3 keV could be fitted by a state whose properties are consistent with a known subthreshold $J^{\pi}=1^-$ level at $E_x=8.039$ MeV. The cause of the $1/v$ cross section near thermal energy could not be determined although the known 2^+ state at 8.213 MeV was found to be too narrow to contribute much to the thermal cross section. Our data are compared to measurements made via the inverse reaction. There are many differences between the two sets of data. The astrophysical reaction rate was calculated from the measured cross section. This reaction plays a role in the nucleosynthesis of heavy elements in nonstandard big-bang models. At big-bang temperatures, the experimental rate was found to be in fair agreement with the rate estimated from the previously known properties of states of ^{18}O in this region. Furthermore, using the available information from experiments, it was estimated that the $^{17}O(n,\alpha)^{14}C$ rate is approximately a factor of 10^3-10^4 times larger than the $^{17}O(n,\gamma)^{18}O$ rate at big-bang temperatures. As a result, there may be significant cycling between ^{14}C and ^{17}O resulting in a reduction of heavy-element nucleosynthesis.

I. INTRODUCTION

The structure of ^{18}O has been studied through many reactions, and a fair amount is known about the energies, spins, and parities of levels near the neutron threshold. However, the widths of the levels are often only approximately known. From previous measurements on ^{7}Be (Ref. [1]) and ^{22}Na (Ref. [2]) we have found (n,p) and (n,α) cross sections near the neutron threshold to be very helpful in elucidating the structure of the compound nucleus which is formed. Except at thermal energy, there have been no reported measurements of $^{17}O+n$ cross sections for energies below a few MeV. In principal, the $^{17}O(n,\alpha)^{14}C$ cross section could be determined from published measurements [3,4] of the inverse reaction. Below a neutron energy of a few hundred keV, however, the rapid decrease of the $^{14}C(\alpha,n)^{17}O$ cross section, together with background from the $^{13}C(\alpha,n)^{16}O$ reaction makes these measurements very difficult; hence, a direct measurement of the $^{17}O(n,\alpha)^{14}C$ cross section in this region is desirable.

Recently there has been much interest in the possibility of synthesizing heavy elements in so-called nonstandard models of the big bang [5–21]. Whereas nucleosynthesis in standard big-bang models [22] effectively stops at $A=7$, it has been speculated that the large density inhomogeneities possible in nonstandard models may lead to the synthesis of elements [10] with mass $A\geq 12$. Presently it is thought that ^{14}C may act as a bottleneck in the path to heavier elements [20]. If this difficulty can be surmounted, however, then the nucleosynthesis can proceed, mainly via a series of neutron captures, through nitrogen and oxygen and on to the heavier elements. However, if the $^{17}O(n,\alpha)^{14}C$ cross section is large enough, then much of the material will be cycled back to ^{14}C, reducing the yield of heavier elements [10]. Using the known thermal cross sections for ^{17}O, and what is known about the properties of states in ^{18}O near the neutron threshold, it is expected that the (n,α) rate will be much larger than the (n,γ) rate at big-bang temperatures. However, some of the total as well as the partial widths of the states in ^{18}O near the neutron threshold are uncertain or undetermined, so a direct measurement of the $^{17}O(n,\alpha)^{14}C$ cross section would be very helpful in reducing the uncertainty in this rate and, subsequently, in reducing the uncertainty in the big-bang calculations.

II. EXPERIMENTAL PROCEDURES

The measurements were performed at the moderated "white" neutron source of the Manuel Lujan, Jr. Neutron Scattering Center (LANSCE) [23] using an apparatus which has been described elsewhere [1], so only the salient features will be mentioned. The data were taken in two parameter mode, pulse height (or alpha energy) versus time of flight (or neutron energy). In this way pulse-height spectra at all energies were measured simultaneously.

The ^{17}O samples were made by anodizing niobium [24] in water enriched to 37.5% in ^{17}O. The thickness of the oxide layer was determined by the applied voltage. Samples ranging from 50 to 350 $\mu g/cm^2$ of Nb_2O_5 were prepared on Nb which was 25.4 μm thick. Most of the measurements were made with a Nb_2O_5 layer which was 200 $\mu g/cm^2$ thick.

Alpha particles from the $^{17}O(n,\alpha)^{14}C$ reaction were detected with a silicon surface-barrier detector which was 10 μm thick by 50 mm^2 in area. Representative pulse-height spectra are shown in Fig. 1. It was necessary to use so thin a detector to reduce the background from neutron interactions in the rather thick Nb target backing to acceptable levels. The main source of background

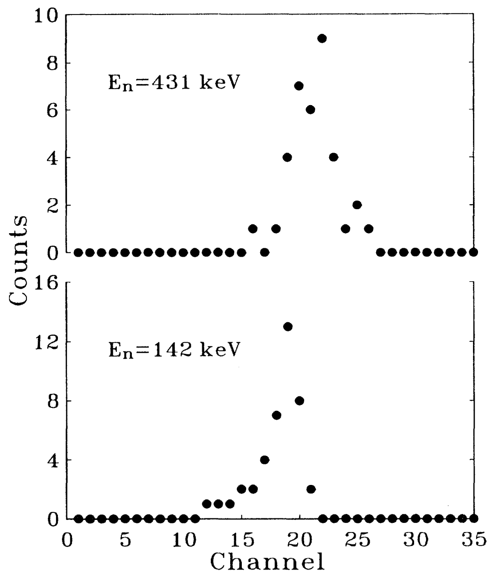

FIG. 1. Representative pulse-height spectra from our measurements. Each spectrum is labeled with the corresponding incident neutron energy at which it was measured.

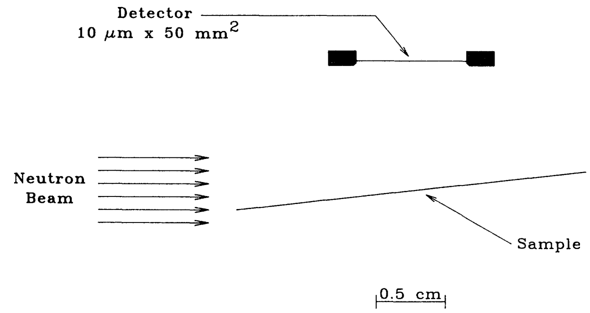

FIG. 2. Schematic diagram of the sample and detector geometry used in our experiment. The neutron beam was collimated to approximately 0.5 cm in diameter. The detector was 10 μm thick by 50 mm^2 in area. It was placed at 90° to the beam at about 1 cm from the center of the sample. The sample was 2.5 cm long by 0.5 cm wide. It was inclined to the incident neutron beam so that its front edge was approximately 0.3 cm lower than its rear edge.

was due to neutrons scattered from the Nb interacting with the boron dopant [via the $^{10}B(n,\alpha)^7Li$ reactions] in the silicon detector. The α_1 group from the ^{10}B reaction was not fully resolved from the α_0 group from ^{17}O. This background was measured by using a blank Nb foil having the same size as the one used for the ^{17}O sample. The peak from the $^{17}O(n,\alpha)^{14}C$ reaction was identified in the pulse-height spectrum by (i) calibrating the energy scale of the spectrum using ^6Li and ^{10}B samples, and (ii) using ^{17}O samples of different thicknesses and observing that the yield of the peak per unit of neutron flux was correlated with the sample size.

In previous measurements [1,2] we have used circular samples of approximately the same diameter (0.5 cm) as the collimated neutron beam. However, due to the relatively small size of both the cross section and the available detector, we used a larger sample to obtain an adequate counting rate. The sample was 0.5 cm wide by 2.5 cm long. As shown in Fig. 2 it was inclined slightly to the incident neutron beam. The detector was placed at 90° to the neutron beam at about 1 cm from the center of the sample. The $^{17}O(n,\alpha)^{14}C$ cross sections were calculated from the alpha particle yields in the detector assuming the cross section to be isotropic. This is probably not a good assumption at the higher energies where the cross section appears to be dominated by non-s-wave resonances [25].

The measurements were made relative to the $^6Li(n,\alpha)^3H$ cross section using a separate ^6Li sample and solid-state detector as a flux monitor. The data were converted from yields to cross sections using the know thermal (n,α) cross sections for ^6Li (Ref. [26]) and ^{17}O (Ref. [27]), and the latest evaluation for the energy dependence of the ^6Li cross section [26]. The resulting cross sections for ^{17}O were corrected for the anisotropy in the $^6Li(n,\alpha)^3H$ differential cross section as explained in Ref. [1]. This correction was much less than 5% at most energies but was as large as 35% for energies near the peak of the ^6Li+n resonance at 250 keV. The necessary $^6Li(n,\alpha)^3$ H differential cross sections for this correction were obtained from Ref. [26].

The data were taken with a time-of-flight channel width of 64 ns. To improve the statistical accuracy below approximately 100 keV, the data were compressed into 50 bins equally spaced on a logarithmic scale in neutron energy. Above approximately 100 keV the data were left in the original 64 ns wide bins to obtain the best possible energy resolution. In the keV region and above, the resolution is dominated by the width of the proton pulse from the Proton Storage Ring (PSR) at LANSCE. The PSR pulse is triangular in shape with a full width at half maximum of 125 ns and a base of 250 ns (Ref. [28]). Nonrelativistically, the time of flight t, in μs, can be calculated from the incident neutron energy E, in eV, using the equation

$$t = 72.3d/E^{1/2}, \tag{1}$$

where $d=7m$ is the flight path distance. Hence, the resolution at the higher energies is given by

$$\Delta E = 2\Delta t E^{3/2}/(72.3d), \tag{2}$$

where $\Delta t = 0.125$ μs. For example, the energy resolution at 100 keV is 15.6 keV; hence, the energy resolution is quite broad at the higher energies in our measurements.

The time of flight to energy calibration was made with the aid of cobalt and uranium filters which had been placed in the neutron beam ahead of the sample position. During this calibration run, the ^{17}O sample was replaced by a ^6Li sample so that a larger counting rate could be obtained for observing the dips in the time-of-flight spectrum due to the filters. Otherwise, the setup was identical for the two sets of runs. Dips in the time-of-flight spectra due to aluminum and manganese in the mercury shutter windows were also used in the calibration.

III. RESULTS AND COMPARISON WITH OTHER MEASUREMENTS

The $^{17}O(n,\alpha)^{14}C$ cross sections resulting from our measurements are shown in Figs. 3 and 4. The representative error bars shown on our data depict the one-standard-deviation relative errors only. The relative uncertainties are dominated by counting statistics. A normalization uncertainty of 2.9% was calculated from the published uncertainties in the ^{17}O (Ref. [27]) and ^{6}Li (Ref. [26]) thermal cross sections.

The shape of the cross section is close to $1/v$ below about 10 eV. Above this there are, apparently, 3 dominant resonances at $E_n \approx 3$, 130, and 250 keV.

Also shown in Fig. 4 are the data of Sanders [3] measured via the inverse reaction. We calculated (n,α) cross sections from the "forward-yield" data of Fig. 5 of Sanders using detailed balance. Below the resonance at $E_\alpha = 2.553$ MeV it was very difficult to obtain reliable data from this figure, and we did not attempt to read the data below 2.39 MeV. Hence, it was not possible to ascertain whether the bump in our data near $E_n = 3$ keV was observed in data from the inverse reaction. The data of Sanders were taken with a target estimated to be 20 keV thick at $E_\alpha = 1.16$ MeV, and the normalization uncertainty is given as 60%. The solid curve in Fig. 4 was obtained by averaging the data of Sanders over our energy resolution. Even with this averaging, the agreement in shape between the two sets of data is not very good. The best agreement is obtained near the peaks of the resonances at $E_n \approx 250$, 800, and 900 keV. On either side of the 250 keV resonance, our data is much higher than that of Sanders, while below approximately 100 keV the opposite is true. The arrows in Fig. 4 indicate the energies at which our pulse-height spectra are shown in Fig. 1. These spectra correspond to energies where the data of Sanders are much lower than our data. Although the statistical accuracy of our data is not very high, the background is very low, and our data indicate that the cross section is about five times larger than the values of Sanders at these energies.

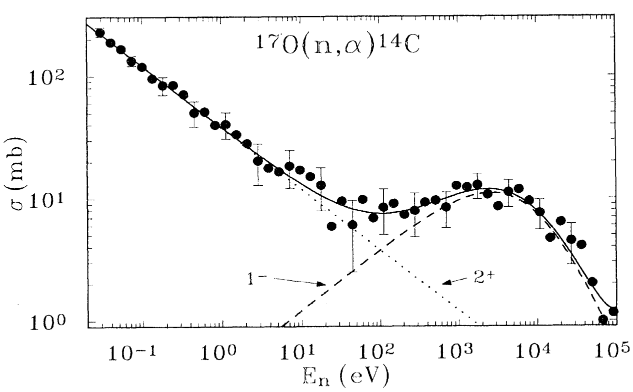

FIG. 3. The $^{17}O(n,\alpha)^{14}C$ cross section from 0.0253 eV to 100 keV. The solid circles are our data. The solid curve is a two-level fit to the data as explained in the text. The dotted and dashed curves are the separate contributions to this fit from 2^+ and 1^- resonances, respectively.

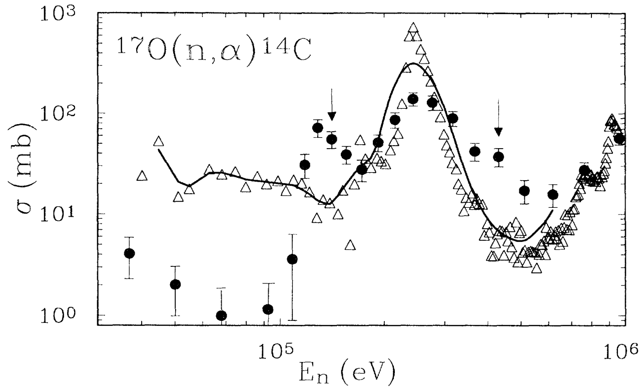

FIG. 4. The $^{17}O(n,\alpha)^{14}C$ cross section from 100 keV to 1 MeV. The solid circles are the data from our measurements. The open triangles are the inverse data of Sanders [3] which we converted using detailed balance. The solid curve resulted from averaging the data of Sanders over the energy spread of our measurements. The two arrows indicate the energies at which our pulse-height spectra are displayed in Fig. 1.

There are several reasons to expect differences between our data and those of Sanders [3] although it is not clear if the magnitude of the difference can be explained. First, the full integrated cross section was not measured in either experiment. From the known spins and parities of the resonances [25] it is expected that angular distribution effects could be important. Second, off resonance the background in the data of Sanders appears to be quite substantial and apparently was not subtracted from the data. For example, there appear to be roughly the same number of counts below threshold as there are above threshold for approximately the first 100 keV. Finally, the dependence of the detector efficiency upon neutron energy was not known in the measurements of Sanders.

It may also be worth noting that the only other reported $^{14}C(\alpha,n)^{17}O$ measurement [4] does not agree well with the data of Sanders below the $E_n = 250$ keV resonance although part of this difference could be due to a different energy resolution in the two experiments.

IV. STRUCTURE OF ^{18}O NEAR THE NEUTRON THRESHOLD

Our limited energy resolution coupled with the fact that we did not measure angular distributions probably precludes the possibility of obtaining much useful nuclear structure information above approximately 100 keV neutron energy, so we will concern ourselves mainly with the region below 100 keV. One interesting feature of the cross section above 100 keV, however, is a bump at $E_n \approx 130$ keV. There is no known level [25] in ^{18}O corresponding to a resonance at this energy. For example, no resonance at this energy was observed in the $^{14}C(\alpha,n)^{17}O$ reaction [3,4]. The bump in our data appears approximately midway between known 5^- and 2^+ (see below) resonances, both of which are apparently too narrow to have been observed in our measurements.

Below 100 keV, the cross section is dominated by a $1/v$ component and a resonance, apparently near 3 keV. There is no known level in ^{18}O corresponding to a laboratory neutron energy of about 3 keV. The nearest known [25] resonance is a 1^- which is bound by 5 ± 2 keV having $\Gamma_{c.m.}<2.5$ keV, and $\Gamma_\gamma=1.07\pm0.22$ eV. Our data below 100 keV can be well fitted by the combinations of a $1/v$ component from a (undetermined) s-wave (2^+) resonance, together with a bound p-wave resonance whose parameters are consistent with the known 1^-. Although the resonance is below threshold, the increase in the p-wave penetrability combines with the decrease in the cross section in the tail of the resonance to produce a bump in the cross section near 3 keV. Furthermore, the calculated bump is not symmetric but, rather, is skewed towards the high-energy side just like the data. We fit the data using simple noninterfering Breit-Wigner shapes,

$$\sigma_{(n,\alpha)}=\pi\lambda^2 g_J\Gamma_n\Gamma_\alpha/[(E-E_0)^2+\Gamma^2/4] \ . \tag{3}$$

The solid curve in Fig. 3 is the sum of a $1/v$ component fitted to the thermal cross section plus a 1^- having parameters consistent with information from other reactions [25] ($E_0=-7$ keV, $\Gamma_\alpha=2300$ eV, and $\Gamma_\gamma=0.9$ eV), and a neutron width $\Gamma_n^0=98$ eV. When the known resonance parameters are kept within the limits from other experiments, the data can be well fitted with $\Gamma_n^0>70$ eV ($\Gamma_n^0=100$ eV is approximately 1% of the Wigner limit). The dotted and dashed curves in Fig. 3 show the separate contributions due to the individual 2^+ and 1^- components, respectively.

If it is assumed that the bump at 3 keV is due to a new resonance, it is not possible to fit both the broad bump near 3 keV and the thermal cross section with a single s-wave (2^+) resonance. To fit the thermal cross section with an s-wave resonance near 3 keV would require a width much narrower (and a peak height much higher) than our data allow. Also, the data cannot be fitted by a combination of a $1/v$ component and a p-wave resonance having $E_0\approx3$ keV while keeping the partial widths within their Wigner limits.

The origin of the $1/v$ component of the low-energy cross section is not clear. The nearest known [25] s-wave resonance is a 2^+ level at $E_n=179$ keV, having $\Gamma_{c.m.}=1.0\pm0.8$ keV, $\Gamma\approx\Gamma_\alpha\gg\Gamma_n$ (Ref. [29]), and $\Gamma_\gamma=0.41\pm0.09$ eV. These parameters limit the thermal cross section to less than about 7 mb, much less than the measured 235 mb. Also, our resolution was too broad to detect a resonance this weak and narrow.

There are apparently no other definite 2^+ levels above the neutron threshold [25] (2^+ assignments have been suggested for the $E_x=8.9$ and 12.04 MeV levels [30]). Below threshold there are three known 2^+ levels, all with fairly large neutron spectroscopic factors measured via the $^{17}O(d,p)^{18}O$ reaction [25] as well as significant α-particle spectroscopic factors as measured with the $^{14}C(^6Li,d)^{18}O$ reaction [30]. Perhaps one or more of these levels is responsible for the thermal $^{17}O(n,\alpha)^{14}C$ cross section and the low-energy $1/v$ cross section. However, the α-particle spectroscopic factors are fairly uncertain and the neutron spectroscopic factors are undetermined for the unbound levels. Also, the possibly large interference effects between the levels have not been measured. Hence, it is not possible at present to determine the origin of the $1/v$ component of the low-energy cross section with much certainty.

V. BIG-BANG NUCLEOSYNTHESIS

The astrophysical reaction rate, $N_A\langle\sigma v\rangle$, calculated from our data is shown in Fig. 5. Also shown is the rate calculated from the data of Sanders [3] shown in Fig. 4, and our data below $E_n=40$ keV (where we were not able to obtain reliably the data of Sanders).

Apparently the $^{17}O(n,\alpha)^{14}C$ rate has never been estimated or calculated and entered in the usual tabulations (e.g., Refs. [31–35]). Hence, we estimated the rate from the previously known properties of ^{18}O resonances near the neutron threshold, plus a $1/v$ component normalized to the measured thermal cross section [27]. The only resonances for which sufficient information is available to calculate the reaction rate are the 2^+ at $E_x=8.213$ MeV, and the 3^- at 8.282 MeV. Judging from the spins, parities, and widths of the other resonances near threshold for which these properties are known, it seems likely that the above two resonances should dominate the reaction rate at big-bang temperatures. However, there are several fairly broad resonances just above our energy range that are known to decay by neutron and alpha emission. Unfortunately, not enough information about the widths, spins, and parities of these levels is known to be able to include them in our estimate.

From the data of Sanders [3], as well as from their own $^{14}C(\alpha,\alpha)^{14}C$ data, Weinman and Silverstein [29] determined that the 2^+ resonance has $\Gamma_n=14\pm9$, $\Gamma_\alpha=1200\pm800$, $\Gamma_{tot}=1280\pm1000$, and $E_0=0.169\pm0.004$. Similarly, the 3^- resonance has $\Gamma_n=830\pm490$, $\Gamma_\alpha=6900\pm400$, $\Gamma_{tot}=8000\pm1000$, and $E_0=0.238\pm0.003$. All quantities are in the c.m. system, the widths are given in eV, and the resonance energies in MeV.

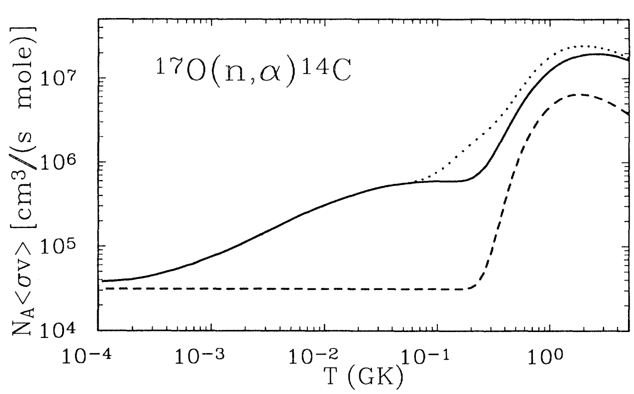

FIG. 5. The astrophysical reaction rate, $N_A\langle\sigma v\rangle$, for the $^{17}O(n,\alpha)^{14}C$ reaction. The solid curve is the rate calculated from our data of Figs. 3 and 4. The dotted curve is the rate calculated from the data of Sanders shown in Fig. 4, together with our data below 40 keV. The dashed curved is the rate calculated from the previously known ^{18}O resonance parameters, plus the thermal cross section.

From these parameters, and the thermal cross section [27], the reaction rate can be calculated [31]. The result is

$$N_A\langle\sigma v\rangle=3.11\times10^4+9.18\times10^5 e^{-(1.961/T)}/T^{3/2}+7.02\times10^7 e^{-(2.759/T)}/T^{3/2}\ \mathrm{cm^3/s\,mole}\ , \quad (4)$$

where the temperature T is in GK. The rate calculated in this way is shown in Fig. 5.

At big-bang temperatures ($T\approx0.3-1.3$ GK), the rate determined mainly from the data of Sanders [3] is approximately 1.4–2.3 times higher than the rate determined from our data, which is, in turn, a factor of roughly 3 times the rate determined from the ^{18}O resonance properties. Because the experimental rates are subject to error due to unmeasured angular distribution effects, and because most of the partial widths used to determine the estimated rate are fairly uncertain, the agreement seems reasonably good. The major exception to this is at low temperatures where the 1^- bound state at $E_x=8.039$ MeV dominates the rate calculated from our data. The estimated rate did not include this resonance because before our measurements its reduced neutron width was unknown.

Because the ^{17}O$(n,\alpha)^{14}$C rate estimated from the previously know resonance parameters is in reasonable agreement with experiment, and because the gamma widths are known, the ratio of the (n,α) to (n,γ) rates calculated from the resonance parameters should be fairly reliable at big-bang temperatures. The thermal (n,γ) cross section is 0.538 ± 0.065 mb (Ref. [36]). The gamma widths [25] Γ_γ are 0.41 ± 0.09 eV for the 2^+, and 0.49 ± 0.13 eV for the 3^-. The resulting reaction rate is

$$N_A\langle\sigma v\rangle_{(n,\gamma)}=71.2+314 e^{-(1.961/T)}/T^{3/2}+5000 e^{-(2.759/T)}/T^{3/2}\ \mathrm{cm^3/s\,mole}\ . \quad (5)$$

As shown in Fig. 6, these estimated rates indicate that the (n,α) rate is approximately a factor of 10^3-10^4 larger at big-bang temperatures than the (n,γ) rate (the ratio at thermal energy is 440). Hence, it seems likely that significant cycling between ^{14}C and ^{17}O may occur in the nonstandard big-bang environment. Previous big-bang calculations have had contradictory results with regards to cycling. For example, Applegate, Hogan, and Scherrer [10] found strong cycling due to the ^{17}O$(n,\alpha)^{14}$C reaction. On the other hand, Kajino, Mathews, and Fuller [21] do not find cycling due to the ^{17}O$(n,\alpha)^{14}$C reaction to be a significant hindrance to the neutron-capture flow to the heavier elements. It is not clear from these papers, however, what values were used for the ^{17}O$+n$ reaction rates. Also, there are significant differences in the two models in terms of the calculated neutron density, the number of fission cycles, and the resulting yield of heavy elements. It would be interesting to know how close the rates used in these calculations are to the rates measured

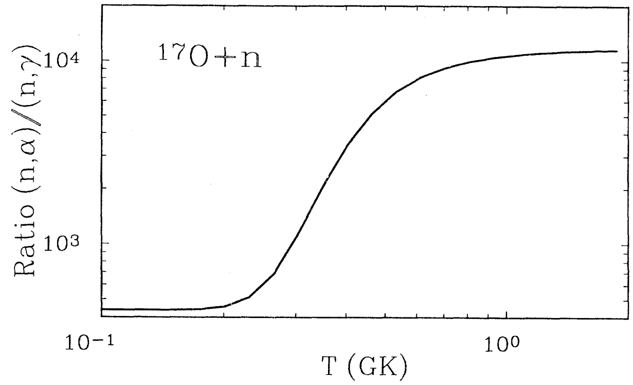

FIG. 6. The ratio of the ^{17}O$(n,\alpha)^{14}$C to the ^{17}O$(n,\gamma)^{18}$O reaction rate. The rates were calculated from the previously known ^{18}O resonance parameters.

and estimated here, and what effect any difference would have on a new calculation.

VI. CONCLUSIONS

Our measurement and analysis of the ^{17}O$(n,\alpha)^{14}$C cross section has revealed an anomaly in the cross section near 3 keV which is caused by a subthreshold 1^- level. Restricting the properties of this level to what is known from previous experiments, the cross section below 100 keV can be well fitted by this 1^-, having $\Gamma_n^0>70$ eV, together with a $1/v$ component fit to the thermal cross section. The origin of this $1/v$ component could not be determined although the known 2^+ level at $E_x=8.213$ MeV was found to be too narrow to contribute much to the thermal cross section.

Our data were compared to data from measurements made via the inverse reaction. Many differences between the two sets of data were found and possible reasons for these differences were discussed.

The astrophysical reaction rate, $N_A\langle\sigma v\rangle$, was calculated from our measured cross sections. At big-bang temperatures, this rate was found to be in reasonable agreement with the rate estimated from previously known ^{18}O resonance parameters. The effect of this rate on the nucleosynthesis of heavy elements in nonstandard big-bang models was discussed. In particular, the available experimental evidence indicates that the (n,α) rate will be about 10^3-10^4 times larger than the (n,γ) rate at big-bang temperatures; hence, cycling between ^{17}O and the ^{14}C "bottleneck" may be important.

ACKNOWLEDGMENTS

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