New $^{13}C(\alpha,n)^{16}O$ Cross Section with Implications for Neutrino Mixing and Geoneutrino Measurements

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Precise antineutrino measurements are very sensitive to proper background characterization. We present an improved measurement of the $^{13}C(\alpha,n)^{16}O$ reaction cross section which constitutes significant background for large $\bar{\nu}$ detectors. We greatly improve the precision and accuracy by utilizing a setup that is sensitive to the neutron energies while making measurements of the excited state transitions via secondary γ-ray detection. Our results shows a 54% reduction in the background contributions from the $^{16}O(3^-, 6.13\ \mathrm{MeV})$ state used in the KamLAND analysis.

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In neutrino mixing, the flavor eigenstates of the active neutrino types $(\nu_e, \nu_\mu, \nu_\tau)$ are related to mass eigenstates via the Pontecorvo-Maki-Nakagawa-Sakata mixing matrix [1]. This phenomenon was the core of the 2015 Nobel Prize [2]. In the three-flavor model, the magnitude of the mass-squared splitting $(\Delta M_{ij}^2 = m_i^2 - m_j^2)$ between neutrino mass states has been determined through a variety of high precision complementary neutrino measurements; $\nu_2 - \nu_3$ mixing was determined from atmospheric [3] and accelerator [4,5] data while $\nu_1 - \nu_2$ mixing was deduced from KamLAND [6] and several solar neutrino experiments [7–9] after applying Mikheyev-Smirnov-Wolfenstein matter enhancement corrections [10,11]. Recent reactor experiments [12–14] have shown the $\nu_1 - \nu_3$ mixing angle to be nonzero, though the sign and the mass hierarchy are still unknown.

The enormous size of these neutrino detectors introduces many sources of background not typically detectable in smaller systems [15,16]. The $^{13}C(\alpha,n)^{16}O$ reaction, in particular, is a major source of background for the Kamioka liquid scintillator antineutrino detector (KamLAND) and other neutrino experiments based on the detection of inverse beta decay (IBD) events $(p + \bar{\nu}_e \rightarrow n + e^+)$. The $^{13}C(\alpha,n)^{16}O$ reaction accounts for more than half of the background rate in the measured neutrino spectrum for KamLAND [16,17]. KamLAND, and similar types of experiments, utilize organic liquid scintillators that contain large amounts of carbon, of which $\sim$1.1% natural abundance is ^{13}C. KamLAND specifically contains 1 kton of organic scintillator [15], of which approximately 10 tons of the active detection material is ^{13}C. Alpha particles from the radioactive decay of low-level actinide contaminants in the liquid, such as ^{210}Po $(E_\alpha = 5.3\ \mathrm{MeV})$, can initiate the $^{13}C(\alpha,n)^{16}O$ (Q value $=$ 2.2 MeV) reaction in the scintillator. Under these conditions, the ^{16}O 0^+ ground state, the first excited (0^+, 6.05 MeV), and the second excited (3^-, 6.13 MeV) final states can all be populated, as shown in Fig. 1. Reaction products from these exit channels can mimic IBD events. These events generate a prompt signal from the e^+e^- decay of the first excited state, followed by a delayed neutron capture signal directly from the reaction [16]. Neutron capture additives such as ^{10}B or ^{6}Li can further reduce this background by requiring a prompt electron-recoil signal followed by a delayed nuclear-recoil signal. However, this does not remove $^{12}C(n,n')^{12}C^*$ or $^{13}C(\alpha,n)^{16}O$ (0^+, 6.05 MeV) reactions, resulting in IBD-like events. Additionally, α-decay events that mimic IBD events have important implications for near-field detection of antineutrinos by limiting the sensitivity of IBD-organic detectors used for reactor monitoring [18].

In order to extract reliable constraints on the two- or three-flavor neutrino mixing angles, θ_{12} and θ_{13}, or to extract geoneutrino spectra, all background sources must be fully characterized and understood. Previously, reactor and geoneutrino detectors [19,20] relied on two sources for the total cross section of the $^{13}C(\alpha,n)^{16}O$ reaction to estimate their backgrounds: the cross section data of Harissopulos

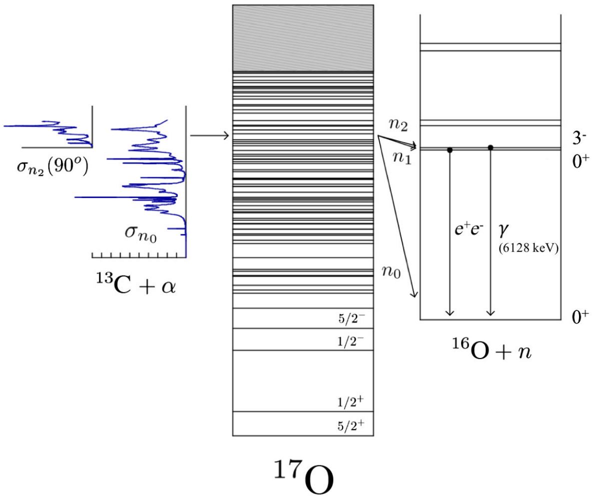

FIG. 1. Level schematic of ^{17}O depicting the pertinent details of the $^{13}C(\alpha, n)^{16}O$ reaction.

et al. [21] and the JENDL/AN-2005 evaluated database [22]. In the case of KamLAND, the cross section data of Harissopulos *et al.* [21] was used for the ground state of ^{16}O, but renormalized by a factor of 1.05 to match *in situ* ^{210}Po^{13}C source measurements [23]. The evaluated cross section to the first excited state at $E_x = 6.05$ MeV was renormalized by a factor of 0.6, and the second excited state data at $E_x = 6.13$ MeV were used with no re-normalization. Since the initial KamLAND result [20], a comparison between reactor on-off data was performed but not before purification of the liquid scintillator, thus a model-independent background is not possible for most of the KamLAND data [24].

In hindsight, the need to renormalize the partial cross sections to match the ^{210}Po^{13}C source measurements is not surprising. The JENDL/AN-2005 evaluation bases its ground state cross section on the inverse $^{16}O(n, \alpha_0)^{13}C$ evaluation of ENDF/B-VI [25], which should be quite accurate. However, due to the lack of experimental data for these transitions, the cross sections to the other final state transitions at $E_x = 6.05$ and 6.13 MeV were determined by scaling the ground state transition by branching ratios determined from a statistical model calculation [22]. However, at these low energies, where individual resolved resonances dominate the cross section, the statistical model approximation is not valid and will result in a poor reproduction of the cross section. As will be shown, the evaluation that resulted deviates from the measured cross section by up to a factor of 4.

As the extracted value for θ_{13} is quite sensitive to the $^{13}C(\alpha, n)^{16}O$ background (cf. Fig. 3 in Ref. [26]), the current work undertook an examination of these cross sections over the energy ranges relevant to KamLAND and other current and future liquid-scintillator-based neutrino experiments [27,28]. This is particularly important as the data of Harissopulos *et al.* [21] was collected with a near 4π moderated ^{3}He counter. ^{3}He counter detection efficiency can be highly energy dependent, as was the case with the detector of Harissopulos *et al.* [21]. Unknown neutron angular distributions and branching ratios to different final states in ^{16}O can lead to very large uncertainties since these directly affect the outgoing neutron energies. At $E_\alpha = 5.3$ MeV, for example, neutron energies vary from $E_n = 4.65$ to 7.29 MeV for the ground state, and $E_n = 0.03$ to 0.36 MeV for the $E_x = 6.05$ MeV excited state transition. The corresponding detection efficiency used in the data of Harissopulos *et al.* [21] ranges from 18.6–24.6% and 38.9–40.0% respectively, a factor of 2 increase in efficiency for the $E_x = 6.05$ MeV state over the ground state, which is much larger than the overall 4% uncertainty claimed in the measurement [21]. This issue was recently highlighted by Peters [29]. The KamLAND analysis assumes a more conservative 11% uncertainty in the cross sections [17]. The $^{13}C(\alpha, n)^{16}O$ reaction has been well studied at lower energies due to its role as the main neutron source for the astrophysical *s* process [30], but for a systematic understanding of the KamLAND and other neutrino experiment backgrounds from this reaction would greatly benefit from measurements of the angular distributions of the neutrons and the branching ratios to the different levels in ^{16}O.

In this Letter, experimental measurements are presented for the individual partial cross sections of the $^{13}C(\alpha, n)^{16}O$ reaction to the ground state and second excited state of the ^{16}O final nucleus. Neutron spectroscopy is used to directly measure the ground state transition, while the second excited state transition is measured via secondary γ rays, owing to the difficulty in detecting the low energy primary neutrons. Measurements of the first excited state transition have not been made as the resulting neutrons are too low in energy for the neutron detectors employed and the state does not produce secondary γ rays (the dominant decay is e^+e^-).

The neutron and γ-ray spectroscopy measurement presented in this work was performed at the University of Notre Dame Nuclear Science Laboratory. The Sta. ANA 5 MV accelerator was used to produce a beam of ^{4}He^{++}, which impinged onto 99% isotopically enriched ^{13}C foils mounted on a tantalum backing. The ^{13}C foils were produced by ACF metals [31] and varied in thickness between 12 and 20 $\mu g/cm^2$. The Ta backing was forced-air cooled and thick enough (0.2 mm) to stop the beam, permitting neutron measurements at 0°. Two deuterated liquid scintillator detectors [32,33] were mounted on a platform at the target location; the first was a deuterated EJ315 (benzene-d$_6$ based) scintillator held at a fixed angle of $\theta_{\rm lab} = 45°$, and the other was a deuterated EJ301D (xylene-d$_{10}$ based) scintillator mounted on a swing arm to permit measurements of neutrons at multiple angles. A single high-purity germanium HPGe γ detector was also mounted at the target location, at $\theta_{\rm lab} = 90°$, to detect γ rays resulting from the de-excitation of the excited ^{16}O final states.

The narrow resonance at $E_p = 992$ keV in the ^{27}Al$(p,\gamma)^{28}$Si reaction [34], as well as calibrated γ-ray sources, were used to determine the HPGe detection efficiency. Neutron detection efficiency was determined using the ^{51}V$(p,n)^{51}$Cr [35–37] and ^{19}F$(\alpha,n)^{22}$Na [38] reactions. For the determination of the target thickness, thick-target yield scans were made of well-known low-energy narrow-resonances in the ^{13}C$(\alpha,n)^{16}$O ($E_R = 1.05$ and 1.34 MeV) and ^{13}C$(p,\gamma)^{14}$N ($E_R = 1.75$ MeV) reactions [39]. Accelerator beam energy calibration was verified using the three low energy resonances in the ^{13}C$(\alpha,n)^{16}$O ($E_R = 1.05$, 1.34, and 1.59 MeV) reaction and the $E_R = 992$ keV resonance in the ^{27}Al$(p,\gamma)^{28}$Si reaction. The beam energy verification depends only on the energy of the resonance and not its magnitude, thus is independent of the normalization of the measured cross section.

An excitation function for the ^{13}C$(\alpha,n_2)^{16}$O reaction was measured at $\theta = 90°$ from $5.2 < E_\alpha < 6.4$ MeV (Fig. 2). Data were recorded at 293 energies throughout the region. At a subset of energies, five-point angular distributions were measured for the ground state neutrons using the EJ301D detector mounted on the swing arm, and on resonances the number of angular steps was increased to 12. The angle integrated cross section is compared to previous measurements in Fig. 3.

We also used spectrum unfolding [33,46] with the neutron detectors, allowing for differentiation between neutrons from different excited states. Pulse shape discrimination (PSD) with the liquid scintillator detectors allowed for clean separation of neutrons and γ rays from the reaction and background sources above a certain energy. This allowed for neutrons resulting from the decay to excited states in ^{16}O to be cleanly separated from those decaying to the ground state n_0 transition. Because of the energy threshold of the PSD, only neutron yields to the ground state transition were extracted. For neutrons de-exciting to the $E_x = 6.13$ MeV excited final state, secondary γ rays detected in the HPGe detector were used. Since the $E_x = 6.05$ MeV state ($J^\pi = 0^+$) cannot decay through γ-ray emission (the predominant decay is through e^+e^- emission) and the emitted neutrons were too low in energy to be detected by the liquid scintillators, the cross section to this transition could not be measured in this experiment. The yields of the 511 keV γ rays in the HPGe were observed to dramatically increase when the beam energy exceeded the threshold for this level, but the cross section could not be extracted accurately from the positron annihilation yields due to uncharacterized background and the long range of the energetic positrons. Since the energy loss of the α beam through the carbon target was small compared to the widths of the observed resonances, cross sections were calculated using a thin-target yield approximation [39].

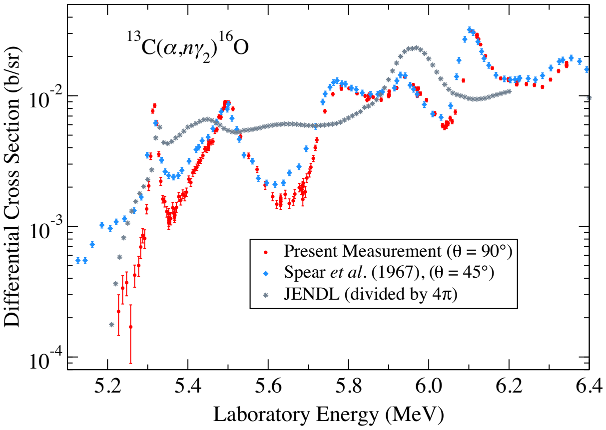

FIG. 2. Comparison of the ^{13}C$(\alpha,n\gamma_2)^{16}$O cross section measured at $\theta_{\rm lab} = 90°$ (this Letter, red circles) and $\theta_{\rm lab} = 45°$ (Spear *et al.* [40], open blue diamonds) to the angle integrated cross section evaluation of JENDL/AN-2005 [22] (gray line) scaled down by a factor of 4π.

In order to compare the present ground-state transition data with previous measurements and evaluations, the measured neutron angular distributions from the current work were subjected to a Legendre polynomial fit to deduce the angle-integrated cross section at each beam energy. The present ^{13}C$(\alpha,n_0)^{16}$O data are compared with the ^{13}C$(\alpha,n)^{16}$O total cross sections of Bair and Haas [41] and Harissopulos *et al.* [21] and with the ^{16}O$(n,\alpha_0)^{13}$C data (using a detailed balance) of Davis *et al.* [42] and Giorginis *et al.* [43] in Fig. 3. The evaluations of the ^{13}C$(\alpha,n_0)^{16}$O

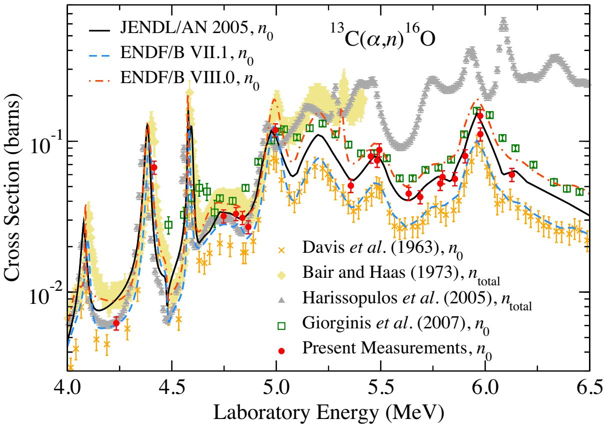

FIG. 3. Comparison of ^{13}C$(\alpha,n)^{16}$O cross section data and evaluations over the energy range of the present measurements. Total cross section data (sum over all final state transitions) are shown for Harissopulos *et al.* [21] (gray triangles) and Bair and Haas [41] (khaki diamond) and ground state transition data for Davis *et al.* [42] (orange cross), Giorginis *et al.* [43] (green open square), and the present measurements (red circles). The evaluated ground state cross sections are shown for JENDL/AN-2005 [22] (black line), ENDF/B-VII.1 [44] (blue dashed line), and ENDF/B-VIII.0 [45] (red dashed-dotted line).

TABLE I. Comparison of estimated $^{13}\mathrm{C}(\alpha,n)^{16}\mathrm{O}$ background events between KamLAND analysis and data from this Letter.

Background source	Interaction	KamLAND [17]	Present work
$^{13}\mathrm{C}(\alpha,n)^{16}\mathrm{O}_{\mathrm{g.s.}}$	$np\rightarrow np$	171.7 ± 18.2	157.5 ± 22.5
$^{13}\mathrm{C}(\alpha,n)^{16}\mathrm{O}_{\mathrm{g.s.}}$	$^{12}\mathrm{C}(n,n')^{12}\mathrm{C}^{*}$	7.3 ± 0.8	6.7 ± 1.0
$^{13}\mathrm{C}(\alpha,n)^{16}\mathrm{O}(0^{+},6.05\ \mathrm{MeV})$	6.0 MeV $e^{+}e^{-}$	15.9 ± 3.3	(26.5 ± 5.5)[a]
$^{13}\mathrm{C}(\alpha,n)^{16}\mathrm{O}(3^{-},6.13\ \mathrm{MeV})$	6.13 MeV γ	3.7 ± 0.7	1.7 ± 0.5

[a] Change reflects omission of the 0.6 scaling factor used in KamLAND [17]. See text for discussion.

cross section are from JENDL/AN-2005 [22], ENDF/B-VII.1 [44], and ENDF/B-VIII.0 [45]. It is immediately apparent that the cross sections of Harissopulos *et al.* [21] are in reasonable agreement with the evaluations below the threshold of n_1 de-excitation ($E_\alpha\approx5$ MeV). Above the threshold, the current data agree well with the JENDL/AN-2005 evaluation, which is between the two more recent ENDF/B-VII.1 and ENDF/B-VIII.0 evaluations.

Due to the use of ^{3}He counters, the measurements of Harissopulos *et al.* [21] were unable to differentiate between neutrons originating from reactions to the ground or excited states. Since the efficiency could not then be properly corrected, this resulted in an overestimation of the total cross section above the excited-state thresholds. This has been discussed recently by Mohr [47] where the correction factors to the data of Harissopulos *et al.* [21] are estimated using the statistical code TALYS. As pointed out by Mohr [47], this gives a rough estimate of these correction factors, but since TALYS relies on a statistical model, for which it is well known that this is certainly not valid in the present case [48], cross section measurements of the individual transitions are required.

The present measurements of the secondary γ rays at $\theta=90°$ can be compared with the data of Spear *et al.* [40] (Fig. 2). Despite different angles of observation, the data present similar resonances and the on-resonance cross sections are quite similar. To approximate the angle integrated cross section, consider the angular distribution as an expansion of Legendre polynomials. First, the expansion is simplified because secondary γ-ray angular distributions can only have even terms, even in off-resonance regions [49]. Second, at $\theta=55°$ the second order Legendre polynomial becomes zero, therefore the differential cross section of Spear *et al.* [40] at $\theta=45°$ is taken as it is the closest to this angle. Finally, given the relative proximity to the threshold and the energy dependence of the Coulomb penetrability, it is expected that higher order terms in the Legendre expansion will be small. Given these considerations, the angle integrated cross section is approximated by multiplying the differential cross section of Spear *et al.* [40] at $\theta=45°$ by a factor of 4π.

Given the approximations noted above, the cross section for the $E_x=6.13$ MeV transition from the present work and that of Spear *et al.* [40] can be compared with the JENDL/AN-2005 evaluation [22] (Fig. 2). While the order of magnitude of the cross sections is similar, the current measurements and those of Spear *et al.* [40], show a quite different resonance structure. The $^{13}\mathrm{C}(\alpha,n\gamma_2)$ cross section for the JENDL/AN-2005 evaluation was calculated by scaling the total cross section by a branching ratio that was determined using a statistical multistep reaction code [22]. As with the corrections made by Mohr [47], the use of a statistical model is not valid in this case as can be seen by the discrepancies in the data.

Of ultimate importance is the effect that an overestimated $^{13}\mathrm{C}(\alpha,n_0)^{16}\mathrm{O}$ cross section has on KamLAND and other measurements. The current results suggest that accounting for this cross section difference results in a decrease in the ground-state contribution to the low-energy background in the KamLAND detector. However, the corresponding increased contribution from the excited states could contribute to an increase in the higher energy background of KamLAND (cf. Fig. 3, Ref. [17]). This contribution can be estimated by a comparison of the ratio of reaction yields between the present data and the KamLAND assumptions. The background contributions resulting from the different partial cross sections of the $^{13}\mathrm{C}(\alpha,n)^{16}\mathrm{O}$ reaction on the KamLAND measurements are summarized in Table I. As mentioned, the scaling factors used by KamLAND were based on fits of a $^{210}\mathrm{Po}^{13}\mathrm{C}$ source measurement to experimental data of Harissopulos *et al.* [21] for the ground state and JENDL/AN-2005 for the excited states. The data of Harissopulos *et al.* [21] can not be used for the ground state cross section above the threshold for population of excited states without corrections, as shown in Mohr [47], due to the unknown contributions of the excited states. Thus the 0.6 scaling of the $^{13}\mathrm{C}(\alpha,n)^{16}\mathrm{O}(0^{+},6.05\ \mathrm{MeV})$ state is no longer justified and should not be used pending further measurements. Table I reflects the contributions of the $^{13}\mathrm{C}(\alpha,n)^{16}\mathrm{O}(0^{+},6.05\ \mathrm{MeV})$ without the 0.6 scaling used by KamLAND. In addition, we find a 14% relative error on the ground state contribution and larger errors for the excited state contributions thus the 10% uncertainty claim in Harissopulos *et al.* [21] for the discovery of geoneutrinos will need to be revisited.

The current work elucidates the significant discrepancies in the $^{13}\mathrm{C}(\alpha,n)^{16}\mathrm{O}$ data used in the analysis of the backgrounds in neutrino oscillation measurements. One immediate need is to extend the current measurements to $E_\alpha \approx 8$ MeV to examine backgrounds from the decay of uranium, thorium, and actinium (and their daughters). These actinides are typically in higher concentration for Gadolinium loaded detectors due to difficulties in radio purification [50] of the Gd additive. Because the data for the second excited state differed from the JENDL/AN-2005 evaluation, a dedicated measurement of the cross section to the first excited state is recommended.

In conclusion, a systematic and high-precision measurement of the $^{13}\mathrm{C}(\alpha,n)^{16}\mathrm{O}$ cross section, including discrimination between the n_0, n_1, and n_2 branches with high-resolution angular distributions, was undertaken. The results indicate that the $^{13}\mathrm{C}(\alpha,n)$ cross sections adopted by the KamLAND Collaboration and similar experiments in estimating detector backgrounds are inaccurate, substantially exceeding the adopted uncertainties at some energies in the prompt energy spectrum. This in turn alters the measurement's sensitivity to the θ_{13} neutrino mixing angle, and we encourage the KamLAND collaboration to assess the impact of these new results.

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