Energy of the 9.17 MeV excited state of ¹⁴N

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The 9.17 MeV excited state of ¹⁴N has been populated using the sharp resonance at 1.75 MeV in the ${}^{13}C(p,\gamma){}^{14}N$ reaction, and the energies of four sequential γ rays deexciting the state to ground have been measured on a scale derived from the known energies of ${}^{56}Co$ lines. An excitation energy of 9171.540(38) keV is determined, and the energies of two other states are found to be 6445.967(26) and 3947.904(17) keV.

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I. INTRODUCTION

In the AURA2 laboratory we have a long-term program whose aim is to measure both the energies and the partial halflives of those 0+ to 0+, T=1 superallowed positron decays whose Ft values may be determined with a precision of better than 0.1%. This aim is particularly difficult to achieve for the energies, as the F value depends roughly on the fourth power of the energy. In addition, the local realisation of the energy unit, the MeV, is not straightforward. To address these problems we have developed the heavy ion source system (HISS), see Ref. [1], which typically aims to determine a 6 MeV positron decay energy by relating it to a 7 MeV proton threshold energy which has been measured with a precision approaching 10 ppm, on an MeV scale which is tied to a Josephson one-volt standard. For example, in Ref. [1] we quote the threshold for the ${}^{38}\text{Ar}(p,n){}^{38}\text{K}^m$ reaction as 7008.52(12) keV.

Because the HISS method, and the precision claimed, are relatively unusual [for example, there are three other measurements of the ${}^{38}\text{Ar}(p,n){}^{38}\text{K}^m$ threshold energy cited in Ref. [1], but none carry ascribed errors of less than 0.6 keV, or establish their own energy units], it would be reassuring to be able to check them against a generally accepted and high precision energy standard. The masses of the light, stable atoms are now known to a precision of better than 1 eV, see Ref. [2], and so the mass difference $[{}^{1}H+{}^{13}C-{}^{14}N]$ may be taken to be [7288.969+3125.011-2863.417]=7550.563 keV with a precision approaching 1 eV. There is an intense, narrow resonance in the ${}^{13}C(p, \gamma){}^{14}N$ reaction at a proton center-of-mass energy E_p of 1.62 MeV, which is to a state in ¹⁴N at an excitation energy E_x of 9.17 MeV. If E_p could be determined by the HISS method, and E_x by summing the energies of the γ rays connecting the excited and ground states, their difference should agree numerically with the mass difference above. It was the aim of the present work to determine E_x with sufficient accuracy that a subsequent measurement of the resonance energy could be used as a test of the HISS system, preferably at a level approaching 10 ppm.

Although the predominant decay path of the ¹⁴N, 9.17 MeV state is via a direct decay to the ground state (91%, see Ref. [3]), there are no γ -ray calibration energies with sufficient precision that can be used in that energy region. A convenient source of calibration γ rays, with energies from

0.8 to 3.5 MeV which have recently been quoted with high precision, see Ref. [4], is ⁵⁶Co, halflife 77 days, and this may be easily prepared in the laboratory. The main decay paths of the 9.17 MeV ¹⁴N state are shown in Fig. 1, which is taken from Ref. [3]. Using the intensity information contained there, together with the results of further exploratory experiments, it was decided to derive the 9.17 MeV energy from measurements of the energies involved in the cascade 2.73 MeV(9%)-2.50 MeV(2%)-1.64 MeV(2%) -2.31 MeV(2%), with calibration energies coming from sources of ⁵⁶Co.

II. EXPERIMENTAL METHOD

The experimental arrangement is shown in Fig. 2. Targets of freshly evaporated carbon, enriched to 92% in ¹³C, and on a 99.995% pure gold substrate 0.125 mm thick, were attached to a water-cooled backing and bombarded with a 4-5 μ A proton beam from the AURA2 electrostatic tandem accelerator. The beam was collimated through two 3 mm diameter holes, 0.7 m apart, upstream of the target, as shown, and its energy was chosen to lie a few keV above the 1.75 MeV resonance so that the resonant state, of width roughly 100 eV, was completely populated as the protons lost energy in their passage through the target.

 γ rays emitted from the target were detected in a 40% Ge detector, housed within a 254 mm × 254 mm NaI suppressor,



FIG. 1. The principal decay paths of the 9.17 MeV state in ¹⁴N. The more intense arrows represent the γ rays discussed in the present work.



FIG. 2. A plan view of the experimental arrangement.

and the geometrical acceptance of the system was defined by a 20 mm diameter lead tube, through a 50 mm lead wall. Gamma rays of interest, of typical energy 2.5 MeV, passing from the target to the detector on a path not along the tube, were attenuated in intensity by a factor of at least 25.

The time resolution achieved for the Ge-NaI coincidence system for 2 MeV γ rays was roughly 30 ns FWHM with typical rates of 40 kHz(NaI) and 2 kHz(Ge), but the time acceptance window was opened to 150 ns to include all real events. Under these conditions a suppression ratio of only 3.6 was attained, largely because the Ge detector had not been specifically designed for a suppression system. Even this, however, was a desirable improvement over a nonsuppressed situation, as the weak gamma rays of interest, with full energies from 1.6 to 2.7 MeV, lay on a continuum compton background from the strong 9.2 MeV line, which was thus reduced.

A feature of these measurements is that the ¹⁴N nuclei which emit the γ rays of interest are recoiling after the nuclear interaction. As there is only one particle in the final state, all the nuclei are initially traveling at a β (=v/c) of 0.4%, in a direction (0 ± 0.25) degrees relative to the beam axis, and the consequences of the ensuing first order doppler effects are not negligible. Indeed, even the relativistic second order effects must be taken into account. Accordingly, for the ¹⁴N doppler affected γ rays to be intercompared with those from a ⁵⁶Co calibration source, the beam-target interaction point must be on the detector axis, and the latter must be at 90° to the beam direction. To enable this, the Ge-NaI system, with its lead collimation and shielding, was mounted on a table on which it could be rotated about the target. With the detector axis defined by the lead cylindrical collimator, this was aligned geometrically using a snug-fitting insert with a conical point, and this method was judged to be reliable to 1°, which subsequent analysis showed to be realistic.

Experience has shown that, for the energies of the γ rays from two sources to be able to be intercompared, the sources must be at the same angle to the detector, and at approxi-

mately the same distance. In the present case, the 56 Co source was a 3 mm disc, taped 7 mm behind the target, on the detection axis. And further, variation in the relative countrates from the sources during the accumulation of a spectrum should be avoided. Here, data was recorded in two hour spectra, and so the rate from the 56 Co did not change appreciably. For the 14 N, care was taken to keep the beam intensity on target, and hence the 14 N countrate, constant to within a few percent.

Analogue pulses from the Ge detector system were digitised in 16384 channels using a ND579 ADC, whose reliability and good generic linearity had been previously attested to. Spectra were taken at four amplifier gains, each of around 4 chan/keV, to sample different parts of the range of the amplifier-ADC combination. Additional spectra were taken of the ⁵⁶Co source on its own, to enable the performance of the system to be studied and parametrized. As will be discussed later, spectra from ¹⁴N using a variety of detector positions differing by known (geometrically established) angles were taken to allow the determination of the mean nuclear recoil speeds for each of the emissions of the four γ rays of unknown energy.

III. DATA ANALYSIS

The data consisted of suppressed Ge spectra, covering an energy range from 0.5 to 3.5 MeV, each in 16 384 channels, at a dispersion of around 0.26 keV/chan. A typical, and relevant full energy peak from 56 Co (at 2.6 MeV) had a FWHM of 18 channels, while the 14 N peak with greatest doppler broadening (at 2.7 MeV) had a FWHM of 22 channels. A basic problem, which we discussed in Ref. [5], was to find an algorithm which enabled a position to be assigned to a peak, and which reliably represented both doppler broadened and nonbroadened peaks over a 1.3–3.2 MeV energy range. In addition, since the spectrum is quite dense, the algorithm should not only assign a position to the peak, but should represent its shape well, including the "tails."

As reported in Ref. [5], despite many algorithms having been used to automate the analysis of γ -ray spectra from Ge detectors (see, for example, Ref. [6] for a critical evaluation of these), none satisfied the criteria discussed above, and so a different approach was developed.

Instead of attempting to represent a peak as an algebraic function Y(x), of the channel number x, a generic peak shape f(x), was adopted by taking an intense, unbroadened peak from the middle of the energy region of interest, and then subjecting it to two degrees of binomial smoothing. Any other peak was then described as $P_1*f[P_2*(x-P_3)]$, in which the parameter P_1 is an amplitude normalizer, P_2 gives a variable width, and P_3 is the peak position.

The success of this approach may be seen in Fig. 3, which shows, on a logarithmic scale, the fits to the principal calibration lines of 56 Co at 1.36, 1.77, 2.03, 2.60, 3.01, and 3.25 MeV in terms of the smoothed shape of the 2.60 MeV line. As in Ref. [6], the displayed residuals are the differences between the data and the fit, divided by the standard errors. Visually the fitting method is seen to be successful and it was tested in a pragmatic way by using the strong 56 Co lines



FIG. 3. Fits to the shapes of the ⁵⁶Co lines at 1.36, 1.77, 2.03, 2.60, 3.01, and 3.25 MeV, using the shape of the 2.60 MeV line, as described in the text. The residuals are the difference between the data and the fit, divided by the square root of the data.

1810.712(16) keV

Hel. 1810.726(4) keV

1963.706(15) keV

Hel. 1963.703(11) keV

2113.113(26) keV

Hel. 2113.092(6) keV

from 1.36 to 3.25 MeV as a calibration in terms of which to determine the energies of four weak ⁵⁶Co lines which lie in the energy region of interest.

In a typical spectrum, twelve strong lines between 1.1 and 3.3 MeV from ⁵⁶Co were represented as described above, and the best straight line fitted to their energies as a function of their positions. The deviations from this straight line are shown plotted in Fig. 4, where the continuous line is the best fit parabola to the points. It obviously represents them well, and this form of description of the nonlinearity of the system had also previously been found to be reliable Ref. [5]. Using four such calibrations, at differing gains as explained, the energies of the ⁵⁶Co lines at 1.81, 1.96, 2.11, and 2.21 MeV were determined, and the results are shown in Fig. 5, where the more precise values given by Helmer, Ref. [4], are also quoted. The comparison seems more than satisfactory for the first three lines, but fails for the fourth at 2.21 MeV, and this is due to interference from the first escape peak of the 2.73 MeV transition from ¹⁴N, the treatment of which will be discussed shortly.



FIG. 4. Calibration of a γ -ray energy spectrum using the lines from ⁵⁶Co, as described in the text. The residuals are in millichannels.

FIG. 5. Four energy determinations of four weak lines from ⁵⁶Co which lie in the energy range of interest, and their comparison with the accepted values from Ref. [4]. The value for the 2.21 MeV line is obviously wrong.

2213.146(23) keV

Hel. 2212.898(3) keV

(E - 1810) keV

(E - 1963) keV

2113) keV 0.15

ய் 0.05

2213) keV

ш 0.05

0.75

0.70

0.65

0.60

0.80

0.75

0.70

0.65

0.60

0.20

0.10

0.00

0.20

0.15

0.10

0.00



FIG. 6. The width parameter for all the lines of 56 Co from 1.06 to 3.27 MeV, and the fit to this in terms of a parabolic function of the line energy. The six points above the line are from first escape peaks.

Because of this form of interference, both in the above case and for the more important ¹⁴N(2.50 MeV)- 56 Co(3.01 MeV) overlap, the behavior of the system was examined further. When the present algorithm is used to fit a gamma peak in a Ge spectrum, it might be hoped that the width parameter P_2 is a smooth function of the γ -ray energy. The value of P_2 , at energies between 1.0 and 3.3 MeV, for the stronger lines of ⁵⁶Co, is shown in Fig. 6, where the dependence is seen to be close to linear, and can be parametrized satisfactorily, as shown, in terms of a parabolic function of the energy. The points which obviously do not lie on the curve are from first escape peaks, which are expected to be broader. The first escape response of the system is explored further in Figs. 7 and 8. In the first the broader peak structure is seen to be entirely consistent, within the error bars, with a P_2 parameter which is increased by a constant multiplying factor of 1.17(2). In the second, the energies of the first escape peaks are shown to consistently differ from the full energies by [511.000-0.365(34)] keV. Finally, Fig. 9 shows the ratio of the amplitudes of the first escape and full energy peaks, which again is a smooth function, practically linear, of the γ -ray energy, and is absolutely quite small because of the suppression system.

IV. RESULTS

The alignment of the detection system for these measurements was critical. If all four of the 14 N γ 's had been emit-



FIG. 7. The multiplying factor for the width parameter for six first escape peaks from 56 Co. The continuous line is the best fit to the points as a constant, independent of the full energy.



FIG. 8. The energies of eight first escape peaks from 56 Co, in terms of the full energies minus 511.000 keV. The continuous line is the best fit to the points as a constant, independent of the full energy.

ted instantaneously and therefore subjected to the maximum doppler effect, the sensitivity of the 9.2 MeV excitation energy to detection angle would have been 0.7 keV/deg. At the outset, it was intended to improve upon the geometrical method of alignment by applying a method which we first introduced in Ref. [7], in which a Ge detector, looking at γ rays from the ¹⁹F($p, \alpha \gamma$)¹⁶O reaction, was rotated about the target until the angle was found at which the measured energy of the 6.1 MeV γ ray agreed with the accepted value. In the present case, the energy of the gamma ray from the first excited state of ¹⁴N at 2.3 MeV has recently been redetermined as 2312.590(10) keV, Ref. [5], and this information was to be used in a similar fashion.

It became obvious, however, that a more efficient and precise procedure was to effect the alignment mechanically, take spectra and extract energies as outlined above, and then *notionally* rotate the detector until the 2.3 MeV energy was correct, adjusting the 1.64, 2.50, and 2.73 MeV energies correspondingly. This methodology seemed to be quite successful, but placed a greater emphasis on the reliability of the value for the 2.3 MeV energy, the realization of which had led to its having been remeasured (Ref. [5]). It also meant that the effective nuclear recoil speeds had to be measured for each of the γ rays.

Although the 90° position for the detection system was



FIG. 9. The amplitudes of eight first escape peaks from ⁵⁶Co, in terms of the amplitudes of the full energy peaks. The continuous line is the best fit to the points as a parabolic function of the full energy.



FIG. 10. The variation of observed energy of the 1.64, 2.31, 2.50, and 2.73 MeV lines from ¹⁴N as a function of the angle of rotation of the detection system. One nominal degree was 0.99 true degrees. (Five such sets of measurements were taken, see text.)

not known exactly, the relative angular position was precisely measurable. So, a series of five independent measurements was performed, in each of which joint ¹⁴N-⁵⁶Co spectra were taken at several angles, and the energy shifts of the four cascade γ rays were determined. One of these is illustrated in Fig. 10, where it can be seen that there is a linear dependence, as expected, and that the value of the coefficient $\alpha = 10^5 (dE/d\theta)/E$ may easily be extracted. Taking all five result sets, and having due regard for the details of the decay scheme in Fig. 1 and for the fact that the maximum possible value of α is 7.5 per deg., the α values for the four transitions at 1.64, 2.31, 2.50, and 2.73 MeV were set at 3.9(3), 3.9(3), 3.9(3), and 7.5(3) per deg, respectively. (Here the "degrees" were nominal, being actually 0.99 true degrees.)

Armed with the parametrization of first escape peaks, the values of α for the 2.31 and 2.73 MeV transitions and the nominal rotations for each run, we could then fit the ¹⁴N(2.73 MeV)-⁵⁶Co(2.21 MeV) composite peaks to extract the energies of the latter. The results are shown in Fig. 11, where the mean is obviously in good agreement with the accepted value and so one can proceed with confidence to the evaluation of the energies of the three ¹⁴N transitions. To illustrate the sizes of the effects of the factors involved, rel-



FIG. 11. Redetermination of the energy of the 2.21 MeV γ ray from ⁵⁶Co. Compare Fig. 5.

evant features in the analysis of the first run are discussed.

The fits to the shapes of the four ¹⁴N γ -ray peaks, using the algorithm described above, are shown in Fig. 12, and seem to be satisfactory. The raw energies of the γ rays, as determined using the calibration procedure described, were 2312.547(20), 1634.999(21), 2497.819(39), and 2725.189(13) keV, respectively. To rotate the system for the correct 2.31 MeV energy, we note that the energy observed detector should be 2312.590(10) - 0.006hv the =2312.584(10) keV, where the second term is the second order doppler shift, which has been evaluated using the measured α parameter. This 37 eV energy shift corresponds to a rotation angle of $0.4(2)^{\circ}$, where, as throughout, the nondoppler shifted 2.3 MeV energy is treated at this stage as absolute. The extraction of the energy of the 2.50 MeV line takes account of a (very small) contribution to the peak of the ⁵⁶Co(3.01 MeV). The rotation gives corrections to the energies of the 1.64, 2.50, and 2.73 MeV lines of 26, 40, and 84 eV, respectively, whose principal uncertainty come from that in the measured 2.31 MeV energy and so must be incorporated later. The corresponding second order doppler shifts are 4, 6, and 25 eV, and the nuclear recoil energies (which can be calculated essentially exactly) are 0.103, 0.239, and 0.285 keV, respectively, and 0.205 keV for 2.31 MeV.

For the four semi-independent sets of runs, the results for the ¹⁴N energy level differences are shown in Fig. 13. The error bars shown do not include contributions related to the 2.31 MeV transition, and this is particularly noticeable in the seemingly slightly self-inconsistent set for the 2.73 MeV transition. These contributions enter in two ways. First, for each set of runs, the effects on the calculated energies of the 1.64, 2.50, and 2.73 MeV lines depend in a correlated way on the extracted position of the 2.31 MeV line, and this is especially important for the 2.73 MeV line for which the mean α coefficient is twice as big as for the others (see above). In addition, no matter whether the final 9.17 MeV excitation energy is obtained as the mean of four determinations of it, or as the sum of four determinations of each of the



FIG. 13. Four determinations of the three excitation energy differences of 1.64, 2.50, and 2.73 MeV in $^{14}\rm{N}.$ (The error bars do not include all contributions, see text.)

FIG. 12. Fits to the 1.64, 2.31, 2.50, and 2.73 MeV lines from ¹⁴N in terms of the 2.60 MeV line from ⁵⁶Co. The fit to the 2.50 MeV peak includes a very small contribution from the first escape peak of the 3.01 MeV γ from ⁵⁶Co.

mean level energy differences, its value will depend strongly but indirectly on the assumed value of the energy of the 2.31 MeV state. The final results to be quoted, with their errors, are the results of detailed calculations which take both these features into account.

There is a last small correction. Although the peak-fitting algorithm deals with the doppler broadened 1.64, 2.31, and 2.50 MeV γ rays satisfactorily, numerical simulations showed this not to be quite so for the 2.73 MeV line, for which the broadening is considerably more pronounced, partly due to its higher energy, but mainly because the nuclear recoil speed at emission is much higher. Simulations indicated that the broadening shifted the peak position 30(10) eV and so this amount should be subtracted from the calculated energy of the transition. It might be thought that a similar, but smaller, shift should apply to the other two transitions, but these are effectively cancelled by the same feature in the 2.31 MeV line which is being used for the notional rotation.

V. DISCUSSION

The most straightforward way of presenting the results is to recommend excitation energies for levels in ¹⁴N at 9171.540(38), 6445.967(26), and 3947.904(17) keV. These are derived from the energy level differences 2725.573(28), 2498.063(20), and 1635.109(13) keV taken with the first excited state energy of 2312.795(10) keV from Ref. [5]. A comparison with the presently accepted energies, from the compilation of Ref. [3], is shown in Table I. These latter, largely drawn from work in Refs. [8] and [9] which also used the

TABLE I. Energies of 14 N levels: comparison between the values of Ref. [3] and the present work.

Present keV	Ref. [3] keV
2312.795(10)	2312.798(11)
3947.904(17)	3948.10(20)
6445.967(26)	6446.17(10)
9171.540(38)	9172.25(12)

calibration lines of ⁵⁶Co, are not updated for the present values of the calibration energies because the assigned errors are substantially larger than the consequent changes. While some disagreement is evident in Table I, the extent is not large enough to warrant concern, except perhaps for the value of the 2.73 MeV energy difference. In the analyses of Refs. [8] and [9], the γ -ray line shapes are assumed to be Gaussian, and no account seems to have been taken of the large doppler effects, particularly for the 2.73 MeV transi-

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tion, other than by alluding to "corrections made for the lifetimes of the states" which, as discussed at length above, are difficult to do *a priori*.

The final recommended energies for excited states in ¹⁴N are therefore 2312.795(10)V, 3947.904(17), 6445.967(26), and 9171.540(38) keV. The mean energy of the γ ray, deexciting the 1.75 MeV resonance directly to ground in the ¹³C(p, γ)¹⁴N reaction, which would be emitted at 90° degrees to the beam direction is [9171.540(38)-0.323 -0.084) keV=9171.133(38) keV, provided that the beam energy and target thickness were chosen so that the whole resonance was populated.

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