go to final states of spin and parity different from the 0^+ of the 2.31-Mev state. Perhaps the most similar allowed transition is the $Q=-3.95$-Mev transition, which leaves the C^{12} in its ground state and the N^{14} in its second excited state (1^+). The cross section for scattering to the 2.31-Mev state is $<1/15$ that for scattering to the 3.95-Mev state. A similar upper limit (6%) for this intensity ratio was obtained by Watters[19] with 31.5-Mev alphas scattered by N^{14}.

It was pointed out by Hashimoto and Alford[20] in a similar problem that the angular momentum and parity conservation rules should be considered. In this case they would inhibit the 2.31-Mev transition by a factor of about 3. It appears then that although a stringent test of the isotopic-spin conservation rule must be sought elsewhere, within the sensitivity of this experiment the rule is not violated.

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Lifetime of the First Excited State of N^{14}†

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The mean life of the first excited state of N^{14} has been measured by nuclear resonant scattering. The $N^{14}(p,p')$ reaction with an N^{14} enriched gaseous ammonia target was used as the source of Doppler-broadened γ radiation. The value of $(7.3\pm1.8)\times10^{-14}$ sec obtained is a factor of two longer than theoretical predictions based on the wave functions used to explain the anomalously slow C^{14} β decay.

INTRODUCTION

THE slowness of the C^{14} β decay seems to be most satisfactorily explained by the chance cancellation of terms in the matrix element.[1–4] Since the electromagnetic decay of the first excited state of N^{14} involves, in the approximation that isotopic spin is a good quantum number, the same nuclear wave functions, a measurement of this decay rate should be of interest. Elliott[3] and Visscher and Ferrell[4] have calculated this rate using wave functions consistant with the β decay. They predict mean lives of 3.75×10^{-14} and 2.88×10^{-14} sec, which may be compared to the extreme j-j coupling prediction of 7.4×10^{-15} sec.

Sherr *et al.*[5] and Thirion and Barloutaud[6] attempted to measure this lifetime by the Doppler-shift method. Neither group was able to find a target dense enough to give a detectable reduction in the Doppler shift of the γ ray. They give upper limits for the mean life of 5×10^{-13} and 3×10^{-13} sec, respectively.

Previous efforts to measure this lifetime by the nuclear resonance fluorescence techniques served only to establish a lower limit of 6.6×10^{-15} sec, as quoted by Warburton and Pinkston.[7] Thus the theoretical values are within the experimental limits, but these limits are so wide as to be of little significance.

The theoretical lifetimes are well within the range of lifetimes previously measured by the resonance fluorescence technique, and one might expect it to be easily measurable. That this is not the case is the result of two unfavorable circumstances. First, the statistical weight factor in the resonance cross section for this 1–0–1 transition is only $\frac{1}{3}$ compared, for example, to 5 for the favorable 0–2–0 case. Second, the most suitable source of the 2.31-Mev γ ray appears to be the $N^{14}(p,p')N^{14*}$ reaction which gives a rather low yield even at proton energies several hundred kev above the $N^{15}(p,n)$ threshold. The presence of neutrons in this case is particularly troublesome because of the 2.22-Mev gamma ray resulting from neutron capture in the hydrogen of hydrogenous materials. Indeed, this measurement was only possible because we were able to obtain from F. H. Spedding nitrogen depleted in N^{15} by a factor of about 30. The low γ-ray yield along

† This research was supported by the U. S. Office of Naval Research.
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with the natural background—specifically a 2.6-Mev γ ray attributed to ThC^{11}—in the concrete of the target room was the limiting factor in our earlier measurements. With the construction of a low-background target room[8] for the Bartol-ONR Van de Graaff accelerator, a measurement of much improved sensitivity became possible.

EXPERIMENTAL METHOD AND DETAILS

Nuclear resonance fluorescence techniques, including the case where the γ-radiation results from a nuclear reaction, have been discussed in a review article by Metzger[9] and in previous publications.[10–15] Only details of special importance will be mentioned here.

The excitation function for the $N^{14}(p,p')N^{14*}$ reaction shows a 82-kev wide resonance at a proton energy of 3.9 Mev and a narrow (<15 kev) resonance at 4.0 Mev.[16] The yield below these resonances is negligible, and the next resonance, at 4.8 Mev,[16] is enough higher in proton energy to give a large increase in neutron background.

Gaseous $N^{14}H_3$ in the cell shown in Fig. 2 of reference 11, was used as the target. Platinum foils of 8 mg/cm^2 nominal thickness were used for the entrance window since they contained fewer neutron-producing contaminants than the tantalum foils used previously. Figure 1, showing the yield curve for the 2.31-Mev γ ray in the region of interest, was obtained with an NH_3 pressure of roughly $1\frac{1}{4}$ cm of Hg in the cell. For the resonance fluorescence studies about 30-cm pressure of $N^{14}H_3$ was used. A yield of about 10^6 2.31-Mev γ rays per μcoul was obtained at the optimum proton energy. The energy region spanned by the target is indicated in Fig. 1.

The NH_3 dissociated under bombardment (presumably into N_2 and H_2) at first rather rapidly, and then at the expected steadily decreasing rate. Unfortunately, the equilibrium pressure was higher than the breaking pressure of the foils, so that after a few hours bombardment with a 4-μa beam it was necessary to replace the gas, even though the neutron-producing contaminants (presumably from outgassing of the cell) had not become unacceptably large. The use of N_2 would have given higher yield, but it was found that the greater

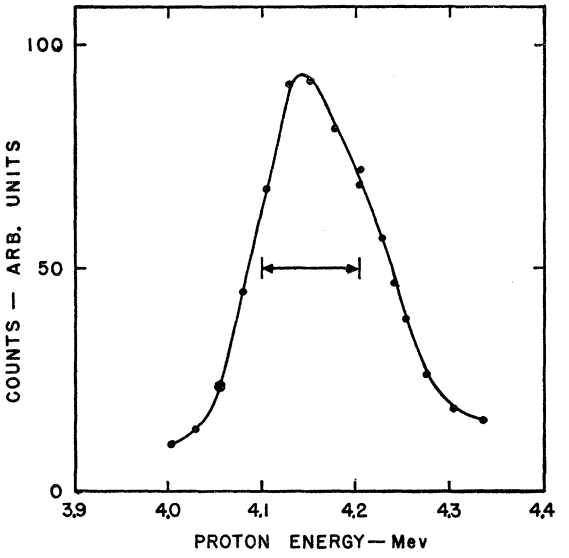

FIG. 1. Excitation function for the 2.31-Mev radiation from the $N^{14}(p,p')N^{14*}$ reaction. The target was gaseous NH_3 less than 10 kev thick for 4-Mev protons. The horizontal arrow indicates the region covered by the target used in the resonance scattering measurements. Because of suspected thickness variations in the entrance window (nominal thickness 220 kev), the curve is probably somewhat less sharp than the true excitation function for the amount of NH_3 used.

efficiency of H_2 in cooling the entrance foil was of importance, so that the net gain was insignificant. The ammonia also had the advantage that it could be transferred by freezing with liquid nitrogen, and that the neutron-producing contaminants it picked up under bombardment could be pumped off when it was frozen.

After considerable investigation it was decided that liquid nitrogen was the most practical scattering material. The construction of the ring scatterer and other details of the experimental arrangement are illustrated in Fig. 2. The scatterer was made about as large as is feasible when the general scale of the experimental arrangement is considered, the scattering volume proper being 33.0 cm i.d. 48.3 cm o.d., and 10.2 cm long and containing 7920 g of liquid nitrogen. The loss by boiling was about 35 g per minute, which required adding nitrogen every 8 min or so, but had the advantage that the gas flow through the vented cork was high enough to obviate any possibility of contamination by condensation of atmospheric oxygen. It was also established that the amount of water vapor condensed was unimportant. The comparison scatterer was an identical container filled with 6120 g of flake graphite. This amount of graphite represents simply the amount which could be easily put in the container, although it is also of the magnitude necessary to match the scatterers for neutron scattering, as suggested by noting that around 1 Mev the total neutron cross section is somewhat higher for carbon than for nitrogen.

As noted in previous publications, when γ radiation

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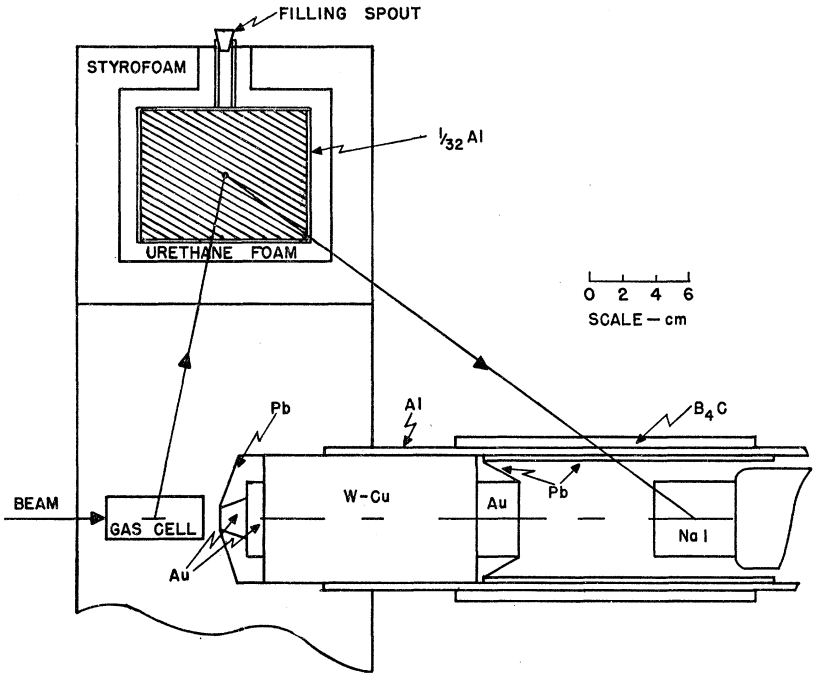

FIG. 2. Scatterer construction and geometry used in the resonance scattering measurement.

is excited in a nuclear reaction such as $N^{14}(p,p')$, the resultant Doppler broadening is very large compared to that necessary to compensate for the nuclear recoil in the emission or absorption of photons. Photons of resonant energy are thus emitted only at approximately 89° to the N^{14*} velocity. Since the inelastic scattering gives N^{14*} only in a forward cone, resonant scattering is expected only when the scatterer is within a cone centered around 89° to the proton beam. If the scatterers are placed outside of this "allowed cone," the scattering angle being kept constant, the matching of the scatterers for effects other than resonant scattering (e.g., neutron scattering) may be verified. In general, the validity of such a measurement depends on the velocity of the nucleus remaining constant until the photon is emitted—in the present case the use of a gaseous target and the observation[5,6] of the full Doppler shift with a solid target is ample assurance that this condition is satisfied.

Also as noted in previous papers, the calculation of the resonant scattering cross section from a scattering measurement requires, for the γ-ray source used, a knowledge of the number of photons per unit energy interval at the resonant energy and the variation of this number as the angle with the proton beam is changed. This function, $N(E_R,\beta)$, depends on both the angular distribution of the recoil nuclei with respect to the beam and the distribution of the γ radiation with respect to the recoil nuclei. The spin of the first excited state of N^{14} is well established as zero, so that it was only necessary to measure the distribution of the recoil nuclei (or the protons). This was done using the variable-angle target chamber, NaI(Tl) proton counter, and gas cell described in reference 11. Spectroscopically pure nitrogen was used as the target gas. The 1.4 mg/cm^2 aluminum windows in the cell give 2.2-Mev γ rays so that integration of the incident proton beam, rather than monitoring the γ-ray yield, was necessary. Measurements were made at five laboratory angles between 35.5° and 145°, and at nine proton energies in the neighborhood of the two resonances. Since only the integral of $N(E_R,\beta)$ over the target thickness is needed, and since calculation of the dependence on β involves an averaging over a range of center-of-mass angles of the proton with respect to the beam direction, these measurements did not have to be made with great care.

RESULTS

The value of $N(E_R,\beta)$ obtained from the angular distribution measurements did not differ much from the value that would be given by an isotropic proton distribution, the largest difference being only 8%, and integration over the scatterer (which covered almost the whole range from the smallest allowed value of β to $\beta=\pi/2$) gave $N(E_R)_{\text{exp}}=0.975\,N(E_R)_{\text{isotropic}}$. As for the inelastic proton distributions, we only comment that they showed a general forward peaking, that there appeared to be interference between the two resonances, and that at the peak of the 3.9-Mev resonance our results are not in disagreement with those of Olness *et al.*[17]

The resonant scattering effect is shown in Fig. 3, which gives the difference between the pulse-height distribution with the nitrogen scatterer and that with the graphite scatterer. The solid curve is the direct

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beam distribution, obtained by exposing the same NaI(Tl) counter, at a distance of 117.5 cm to the direct γ radiation from the target. The rather poor statistics reflect the smallness of the resonance effect, and indicate why a self-absorption measurement was not attempted in this case. Taking the sum of counts from 36 volts to 55 volts as representing the resonance effect, we get 2.13 ± 0.36 counts per 10^5 monitor counts. With the scatterers in a position that corresponds to interchanging the counter and source, none of the γ radiation incident on the scatterer is of resonant energy, as mentioned above. The nitrogen-graphite difference is then -0.09 ± 0.40, giving a net effect of 2.22 ± 0.54 per 10^5 monitor count. The direct beam measurement gives 4.02×10^4 per 10^5 monitor count. It may also be pointed out that the equivalence of the two scatterers for nonresonant effects is supported by the observation that the difference in counting rates for energies greater than 2.31 Mev, for both geometries is -0.33 ± 0.86 per 10^5 monitor count. Since the source itself gives essentially no higher energy γ rays, any difference here would indicate a neutron effect.

The calculation of the resonant scattering cross section and thus of the natural width of the level from the information given requires (in addition to the consideration of such things as the electronic, nonresonant, attenuation of γ radiation in matter) a correction for the self-absorption in the scatterer, i.e.,—the removal of photons of the resonant energy from the beam striking a scatterer element by resonant scattering in elements already traversed. This depends not only on the scattering cross section but also on Δ, the Doppler width of the absorption line due to thermal motion of the scattering nuclei. The value of 2.34 ev used for Δ is that for atomic nitrogen at 77°, i.e., no correction for chemical binding[18,19] has been applied. Since the average self-absorption correction in the present case is only about 5%, any change in Δ will have only a small effect on the final result.

We get then, for the natural width of the first excited state of N^{14}, $\Gamma=(8.6\pm2.1)\times10^{-3}$ ev, which corresponds to a mean life of $\tau=(7.3\pm1.8)\times10^{-14}$ sec. The error given is the same as the statistical error in the data, the other experimental errors being much smaller.

DISCUSSION

The experimental mean life for this transition is longer by a factor of two than the longest of the theoretical predictions given in the Introduction, a difference well outside the experimental error. The matrix element for this $M1$ transition (see, e.g., Sherr *et al.*[5]) contains both a spin-dependant and an orbital part. Cancellation in the C^{14} β-decay matrix element results in the vanishing of the spin part of the γ-decay matrix element. Our experimental result imposes limitations on the intermediate coupling coefficients of the ground and excited state wave functions for N^{14}. For example, a value for the matrix element lying within our experimental error can be reproduced only if the amplitude of the S component of the ground-state wave function is limited to the values either below 0.05 or between 0.4 and 0.6.

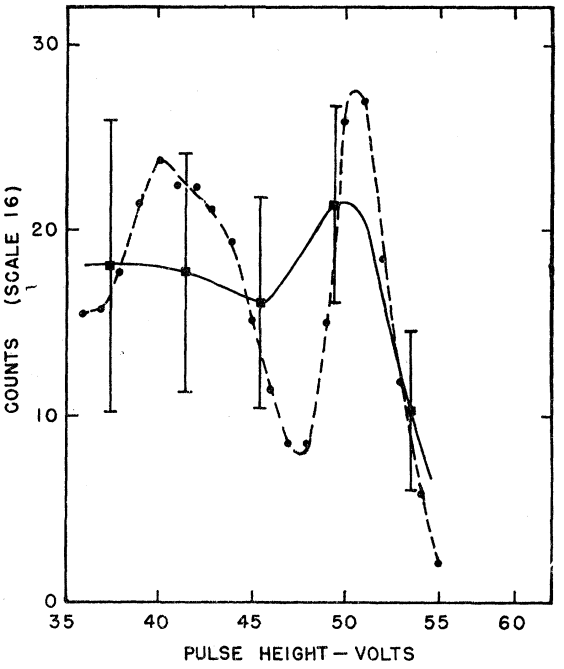

FIG. 3. Pulse-height distribution of the resonance scattered radiation. The individual points were obtained as the difference between the liquid nitrogen and graphite scatterers. The dashed curve gives the pulse distribution of the primary radiation.

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