Mass of ^{64}Ge and Its Role in Nucleosynthesis*

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The mass of the $N = Z$ nucleus $^{64}_{32}$Ge has been measured by determining the positron end-point energy from the prominent decay to the 427-keV state in ^{64}Ga. The ^{54}Fe(^{12}C, $2n$)^{64}Ge reaction was used to produce ^{64}Ge, at a bombarding energy of 36 MeV. Off-line β^+-γ coincidences were studied, as well as the half-life of the 427-keV transition. The half-life was observed to be 70 ± 7 sec, in agreement with a previous measurement. The observed end-point energy was 2.96 ± 0.25 MeV, leading to a total decay energy of 4.41 ± 0.25 MeV, and a mass excess of -54.43 ± 0.25 MeV for ^{64}Ge. Implications for nucleosynthesis and the solar system abundance of ^{64}Zn are discussed.

I. INTRODUCTION

Recently, calculations of nucleosynthesis in supernova models involving detonation of a ^{12}C core have been made by Arnett, Truran, and Woosley.[1] By allowing the nuclear reaction network to extend beyond the Ni isotopes to include nuclei as heavy as ^{66}Ge, they were able to obtain excellent agreement with observed solar system abundances for elements in the iron group. In particular, the elements on the high-Z side of the iron peak (Co, Ni) are produced in such a model.

One of the important nuclei[2] involved in the reaction network is $^{64}_{32}$Ge, an "α particle," or even-even $N=Z$, nucleus. The α-particle nuclei play a major role in the explosive nucleosynthesis of elements up to the mass-60 region, because of their relatively high stability, and also because the material being processed tends to have nearly equal numbers of protons and neutrons. These nuclei are mainly involved in a series of (α, γ) radiative capture reactions [or equivalently, a series of $(\alpha, p)(p, \gamma)$ sequences], which are usually thought to be ineffective beyond ^{56}Ni. During explosive nucleosynthesis at high temperatures, the possibility exists that such a capture chain could also produce significant amounts of heavier nuclei, such as ^{60}Zn and ^{64}Ge. The final abundances for these nuclei are determined by a partial equilibrium, in which important parameters are the binding energy of the last α particle and the freeze-out temperature (the temperature at which the nuclear reactions essentially cease). The participation of ^{64}Ge in this process is of interest because of the high solar system abundance of ^{64}Zn, which is not completely explained by s-process neutron capture.[3]

Until recently the isotope ^{64}Ge had not been observed. It was predicted to be particle stable by Garvey *et al.*,[4] who also predicted its mass. Assuming a ground-state spin of 0^+, ^{64}Ge is expected to decay by positron emission to 1^+ states in 2.6-min $^{64}_{31}$Ga. Using the mass excess calculated by Garvey *et al.*[4] of -54.03 MeV, and the known value of the ^{60}Zn mass excess,[5] the Q value for the ^{60}Zn$(\alpha, \gamma)^{64}$Ge reaction is predicted to be +2.26 MeV. This is also the binding energy or separation energy S_α of the last α particle in ^{64}Ge.

Robertson and Austin[6] (henceforth referred to as RA) have reported the observation of ^{64}Ge. They give a value of 63.7 ± 2.5 sec for the half-life, revised slightly from their published value, but they did not measure the mass.

In this article we confirm the observation of ^{64}Ge, and report measurements of its mass and half-life. The astrophysical implications of the observed mass are discussed.

II. EXPERIMENTAL METHOD

The isotope ^{64}Ge was produced via the ^{54}Fe(^{12}C, $2n$)^{64}Ge reaction at 36-MeV bombarding energy, using the ^{12}C^{4+} ions from the MP tandem accelerator at the Brookhaven National Laboratory (BNL). The target, enriched to 92% in ^{54}Fe, was evaporated on a 0.038-cm gold backing, and was about 470 μg/cm^2 thick. Using the BNL rabbit facility, the target was cyclically bombarded in vacuum for 60 sec, and pneumatically transferred into another room where a 2.5-cm-deep × 4.6-cm-diam NE102 plastic scintillator and a 60-cm^3 Ge(Li) detector were used to detect positrons and

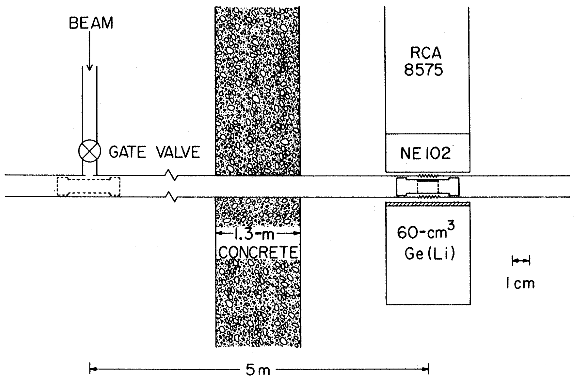

FIG. 1. Experimental configuration for off-line positron-γ coincidence measurement. Not shown are Pb shielding blocks surrounding both detectors.

decay γ rays in close geometry. See Fig. 1 for the experimental configuration. The sensitivity for low-energy γ rays was hampered somewhat by the presence of the gold backing between the target and the Ge(Li) detector, but the most intense transition from ^{64}Ge, at 426.9 ± 0.3 keV, was easily seen. A programmable sequence timer driven by a crystal oscillator was used to time the various functions in the timing cycle. For both the half-life and the mass measurements, counting began 15 sec after the end of the bombarding period. This waiting period allowed short-lived impurities to decrease before counting began.

A. Half-Life Measurement

RA observed five γ transitions which they ascribed to the decay of ^{64}Ge: 128.2 ± 0.2, 384.1 ± 0.3, 427.0 ± 0.3, 667.1 ± 0.3, and 774.5 ± 0.3 keV. For the present half-life measurement, five 50-sec counting periods were used, and 4096-channel γ spectra were accumulated for each time "bin." A pulser signal was injected into the Ge(Li) preamplifier to provide a dead-time normalization. Figure 2 shows part of the γ spectrum in the first 50-sec time bin. Of the five γ rays from ^{64}Ge decay, only the 426.9 ± 0.3-keV γ ray was present with sufficient intensity to be seen. The other γ rays were obscured by contaminant peaks or high background. Other prominent transitions in the spectrum are lines from the decay of 2.6-min ^{64}Ga, which was copiously produced by the ^{54}Fe(^{12}C, pn)^{64}Ga reaction, and lines from 3.4-h ^{61}Cu, arising from the ^{54}Fe(^{12}C, αp)^{61}Cu reaction. The run had a duration of 21 h.

To extract the yield of the 426.9-keV γ ray, the five spectra were each compacted by a factor of 2, and smooth background lines were drawn under the peaks of interest by eye. The top three compacted channels were used to represent the yield. The resulting relative yields were corrected for a contribution from 2.27-h ^{66}Ge at 427.8 keV.[7] The magnitude of this correction was obtained from the intensity of the ^{66}Ge line at 381.9 keV; it ranged from 1.4% for the first time bin to 8.6% for the fifth time bin. The effects of differential absorption by the gold backing, unequal detector efficiencies at 382 and 427 keV, and differing line widths at 382 and 427 keV were taken into account.

In contrast with RA no correction for a transition in the decay of ^{64}Ge at 427.1 keV was applied to the ^{64}Ge yield. Konijn *et al.*[8] reported such a transition with an intensity of 4% of the prominent 992-keV γ ray. Mann, Tirsell, and Bloom[9] gave an upper limit for a 427-keV transition of <0.8% of the 992-keV transition. The observed yield of 426.9-keV γ rays in the present experiment totaled at most 1% of the observed 992-keV intensity, but had a much shorter half-life. In a search for a 427-keV transition in the decay of ^{64}Ga formed

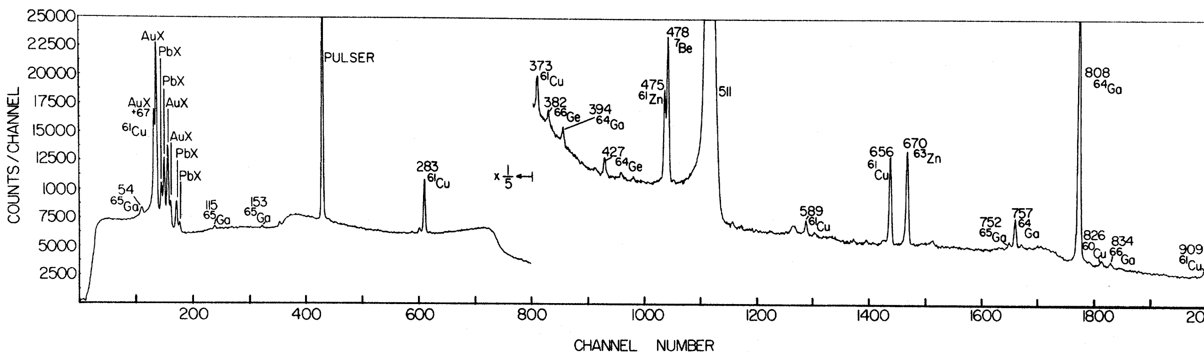

FIG. 2. Part of the γ spectrum for first 50-sec time bin. The 478-keV peak appeared even in the absence of the rabbit, and is attributed to ^{7}Be contamination of the Pb shielding blocks surrounding the detectors. γ-ray energies are given to the nearest keV.

by ^{64}Zn(p, n)^{64}Ga, Davids *et al.*[10] obtained an upper limit for a 427-keV transition in the decay of ^{64}Ga of $(0.9\pm0.9)\times10^{-4}$ per 992-keV transition. For this reason, no correction to the ^{64}Ge yield due to ^{64}Ga was applied.

Another possible correction is from a 427-keV transition following the decay of 38.9-min ^{70}Se.[11] This isotope could be formed by the (^{12}C, $2n$) reaction on a ^{60}Ni target impurity. However, the ^{60}Ni impurity comprised less than 0.02% of the ^{54}Fe target, and thus no correction to the ^{64}Ge yield was necessary.

Figure 3 shows the compacted three-channel yield of 426.9-keV γ rays as a function of time, corrected for ^{66}Ge. Uncertainties are statistical, and reflect a low peak-to-background ratio. The straight line represents the fit obtained from a nonlinear least-squares analysis, and results in a half-life for this transition in the decay of ^{64}Ge of 70 ± 7 sec. This uncertainty includes the least-squares standard deviation, as well as a contribution due to uncertainty in the value of the subtracted background. The result agrees with the value of 63.7 ± 2.5 sec from RA.

As a consistency check, the half-life of the prominent 992-keV transition from the decay of ^{64}Ga was also extracted. The result was a half-life of 157.4 ± 0.7 sec, in excellent agreement with the value of 159 ± 2 sec obtained by Moss *et al.*[12]

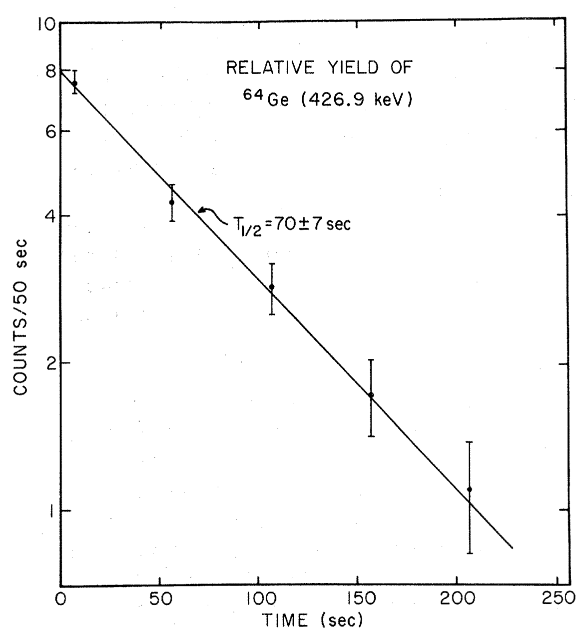

FIG. 3. Relative yield of the 426.9-keV transition as a function of time. The straight line represents the nonlinear least-squares fit for a half-life of 70 sec. The yield has been corrected for a small contribution from ^{66}Ge.

B. Mass Measurement

The decay scheme of ^{64}Ge as reported by RA is shown in Fig. 4, By measuring the energy of the positron in coincidence with the 427-keV γ ray, one can deduce the mass of ^{64}Ge. No γ rays from higher states fed by β^+ decay have even been observed to cascade through the 427-keV state, so that, as far as is presently known, there is a unique β^+ transition in coincidence with the 427-keV γ ray.

Figure 1 shows the β^+-γ coincidence-detection apparatus. Positrons leaving the target traversed a 0.025-cm-thick Be window before entering the detector. Conventional electronic modules were used with a time gate 16 nsec wide for the fast coincidence requirement, allowing a dynamic range from 300 to 1600 keV for γ rays. The β^+ counting rate was limited to 20 000 per sec, maintaining the reals-to-randoms ratio greater than 800 : 1. Bipolar pulses 600 nsec wide were used for the linear β^+-ray signals to minimize pileup. The β^+-ray spectrum in coincidence with γ-ray pulses in the top four channels of the 427-keV peak were stored in one section of an on-line computer. Digital gates 20 channels wide were set on either side of the 427-keV peak, and the β^+ spectra in coincidence with γ-ray pulses within these gates were also stored separately. These latter two spectra served as a measure of the background β^+ spectrum in coincidence with the γ-ray continuum underlying the 427-keV photopeak. The positions of the three digital gates were checked several times during the coincidence mea-

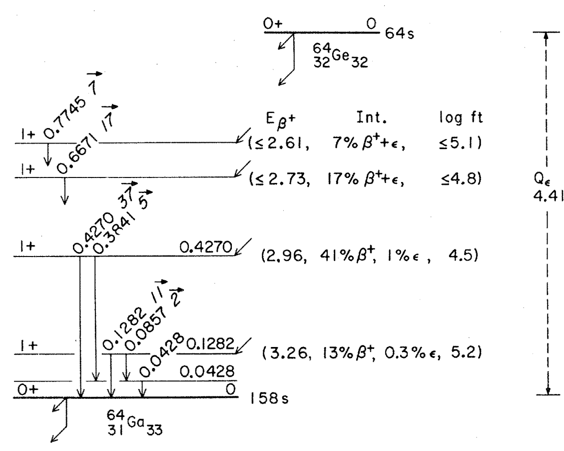

FIG. 4. Decay scheme of ^{64}Ge, taken mainly from RA. The β^+-ray energies and logft values have been recomputed, using the present measured mass. The average of the present half-life and that of RA is 64.4 ± 2.4 sec. The half-life shown for ^{64}Ga is an average of the present value and that of Ref. 14. All energies are in MeV.

surement to ensure that the photopeak gate was properly positioned.

For the coincidence work, data were stored for two consecutive 60-sec time bins starting 15 sec after the end of a 60-sec bombardment. The target was replaced every 3–4 h in a total run of 20 h, to reduce long-lived activities. The resulting two background spectra from the 20-channel-wide gates on either side of the 427-keV peak were identical within statistics, and one tenth of their sum was subtracted from the β^+ spectrum in coincidence with the four-channel-wide gate. This was done for both time bins, and the results had statistically the same shape for each time bin. The corrected results from both time bins were added together and the sum spectrum was compacted by a factor of 4. This spectrum is shown in Fig. 5.

The solid curve in Fig. 5 is a shape fit to the data, and was obtained in the following way. Immediately after the completion of the coincidence work, the gain of the plastic scintillator was determined by storing the spectra due to ^{88}Y and ^{228}Th γ rays. This rough electron calibration indicated a positron end-point energy for the 427 ^{64}Ge transition of close to 3 MeV. The same detection system was then moved to the 3.5-MeV BNL Van de Graaff accelerator, where positron

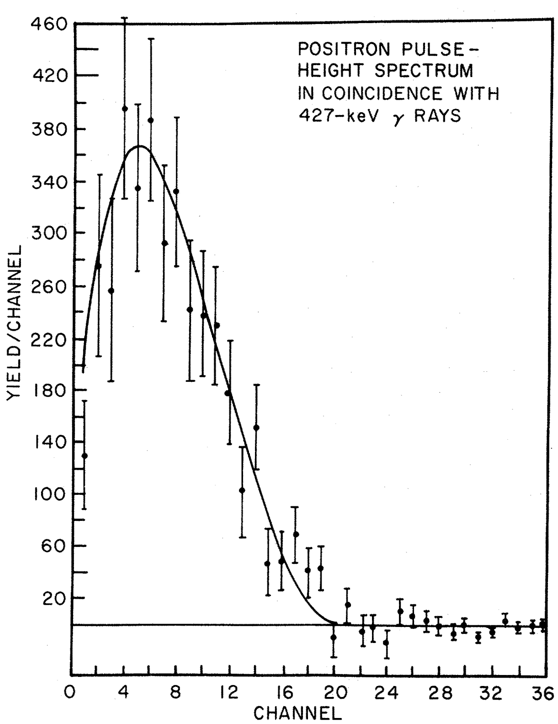

FIG. 5. Spectrum of positrons in coincidence with 426.9-keV γ rays. The curve represents the spectral shape deduced for a positron end-point energy of 2.96 MeV.

calibration sources of 66-sec ^{17}F [from ^{16}O(d, n)^{17}F] and 7.23-sec ^{25}Al [from ^{24}Mg(d, n)^{25}Al] were produced.

During the measurement of a positron spectrum, there exists the possibility of detecting Compton electrons from annihilation quanta, which could then sum with the positron pulse and distort the spectrum. It was therefore necessary to determine the response of the detector to positrons under conditions similar to those encountered in the experiment. ^{17}F and ^{25}Al are ideal calibrations, since they both have essentially (>99%) a single β^+ transition[13, 14] of well-known maximum energy. In addition, their respective end-point energies of 1737 and 3257 keV[5] conveniently straddle the preliminary 3-MeV estimate of the end-point energy for the ^{64}Ge β^+ decay under study.

The gain of the detection system and the zero level of the pulse-height analyzer were then adjusted so that the Compton edges from ^{88}Y and ^{228}Th sources matched to within $\frac{1}{2}$% in channel number those values found just after the coincidence work. A tantalum oxide target with 300 μg/cm^2 of ^{16}O was then bombarded with 2.2-MeV deuterons, and carried to the detection system where ^{17}F positron spectra were separately stored for four successive 60-sec time bins. After a delay of 10 min, four more 60-sec spectra were stored. After dead-time corrections were made, the spectra were corrected for a very small long-lived background. Care was taken to count at nearly the same rate as encountered during the coincidence work, and the geometry was reproduced almost exactly to automatically compensate for the energy loss in the Be window on the rabbit system, the gold backing for the ^{54}Fe target, and nearby lead shielding.

The ^{25}Al activity was made by bombarding natural Mg foil 0.012 cm thick with 3-MeV deuterons, carrying the foil to the remote counting station and storing spectra in many successive time bins. Small corrections for ^{17}F and ^{27}Mg were made. It is possible at this energy for the ^{25}Mg(d, n) reaction to make ^{26}Al, but a time analysis of the spectra showed that the ^{26}Al yield was very small compared to the ^{25}Al activity. ^{25}Al and ^{26}Al have β^+ end points of 3257 and 3212 keV, respectively.[5] This fortunate near coincidence made it unnecessary to be further concerned with the small ^{26}Al activity. An end point of 3250 ± 10 keV was chosen for the resulting ^{25}Al spectrum. A further correction of 10 keV was made because of the thickness of Mg through which the β^+ rays had to pass.

The ^{17}F and ^{25}Al spectra were taken at different counting rates to observe the rate effect on the β^+ shapes. This effect was small, and the calibration counting rate was close enough to the rate used during the coincidence work that the uncertainty incurred was negligible. The ^{17}F and ^{25}Al spectra were then normalized to the same height and the solid curve shown in Fig. 5 was constructed by interpolating between the ^{17}F and ^{25}Al shapes. The result for ^{64}Ge is very close to the shape for ^{25}Al, since its energy is only 9% lower than that of ^{25}Al. The normalized Fermi functions for ^{25}Al and ^{64}Ge are within 1.4% of each other for all channel numbers ≥ 3 in Fig. 5. The first two channels were not used in the shape-fitting procedure. From this fit an end-point energy of 2.96 ± 0.25 MeV is obtained, indicating a total decay energy for ^{64}Ge of 4.41 ± 0.25 MeV. A $\log ft$ of 4.53 ± 0.17 is deduced for the ^{64}Ge β^+ transition under study.

To obtain the mass excess for ^{64}Ge from its total decay energy, the mass excess of ^{64}Ga is needed. This has recently been measured by Davids, Matthews, and Whitmire,[15] using thresholds in the $^{64}Zn(p, n\gamma)^{64}Ga$ reaction. Using their value for the ^{64}Ga mass excess of -58.836 ± 0.006 MeV, the mass excess for ^{64}Ge is deduced to be -54.43 ± 0.25 MeV. This number is to be compared with the calculated value of -54.03 MeV.[4] The experimental mass excess is smaller by 0.4 MeV, indicating that ^{64}Ge is more tightly bound than was previously expected. The separation energy S_α of the last α particle is 2.66 ± 0.25 MeV.

III. ASTROPHYSICAL IMPLICATIONS

In their calculations, Arnett, Truran, and Woosley[1] studied nucleosynthesis occurring in the core of an intermediate-mass star ($4 < M/M_\odot < 9$) which has undergone explosive ignition of the $^{12}C + ^{12}C$ reaction. The detonation wave so produced causes a rapid rise in temperature (to above 6×10^9 °K), setting up a complete nuclear statistical equilibrium. After the passage of the detonation wave, the material expands and cools, passing through a series of equilibrium configurations until freeze-out temperatures for the various nuclear species are reached.

The elements on the high-Z side of the iron peak are primarily formed in the lower-density outer region of the core. This is due to the high abundance of helium, which enables the α-capture chain to proceed as far as ^{64}Ge, and possibly further. An approximate quasiequilibrium between $^{56}Ni(\alpha,\gamma)^{60}Zn(\alpha,\gamma)^{64}Ge$ is formed, finally freezing out at a temperature of about 2.5×10^9 °K.

This partial equilibrium requires that the number densities of ^{64}Ge, ^4He, and ^{60}Zn be related by

$$N(^{64}\mathrm{Ge}) = \mathrm{const} \times N(^{60}\mathrm{Zn}) N(^4\mathrm{He}) T^{-3/2} \exp(S_\alpha/kT),$$

where k is Boltzmann's constant and T is the temperature in degrees Kelvin.

The ^{64}Ge so produced rapidly decays to ^{64}Zn through ^{64}Ge after the explosion. In the original calculations which used the Garvey *et al.*[4] estimate for S_α, the amount of ^{64}Zn produced by the explosion normalized to ^{56}Fe totalled only about 1% of the solar system abundance. Using the experimental value for S_α, a straightforward use of the above relation yields a larger production of ^{64}Ge, as much as a factor of 10 greater. This estimate is confirmed by preliminary calculations of Arnett and Woosley,[16] incorporating the experimental S_α, which show that 20% or more of the solar system abundance of ^{64}Zn can be produced as ^{64}Ge in such an explosive event. These results are in disagreement with those of RA, who concluded that α-capture reactions play no role in the nucleosynthesis of ^{64}Zn.

Because the amount of ^{64}Ge produced in this type of explosive event is significant (mass fractions of 3×10^{-4} or greater), it is reasonable to expect that further α-capture reactions might carry the flow of nucleosynthesis in regions of high helium density to even higher masses. Experimental and theoretical studies to investigate this possibility are presently under way.

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[1]W. D. Arnett, J. W. Truran, and S. E. Woosley, Astrophys. J. 165, 87 (1971).

[2]W. D. Arnett, Invited Paper, Houston Meeting of the American Physical Society Division of Nuclear Physics, 1970.

[3]D. D. Clayton, W. A. Fowler, T. E. Hull, and B. A. Zimmerman, Ann. Phys. (N.Y.) 12, 331 (1961).

[4]G. T. Garvey, W. J. Gerace, R. L. Jaffe, I. Talmi, and I. Kelson, Rev. Mod. Phys. Suppl. 41, S1 (1969).

[5]A. H. Wapstra and N. B. Gove, Nucl. Data A9, 265 (1971).

[6]R. G. H. Robertson and S. M. Austin, Phys. Rev. Letters 29, 130 (1972); and private communication.

[7]F. W. N. de Boer, E. W. A. Lingeman, R. van Lieshout, and R. A. Ricci, Nucl. Phys. A158, 166 (1970).

[8]J. Konijn, R. van Lieshout, J. P. Deutsch, and L. Grenacs, Nucl. Phys. A91, 439 (1967).
[9]L. G. Mann, K. G. Tirsell, and S. D. Bloom, Nucl. Phys. A97, 425 (1967).
[10]C. N. Davids, A. Boudreaux, R. C. Pardo, L. A. Parks, and D. P. Whitmire, private communication.
[11]G. G. J. Boswell and T. McGee, J. Inorg. Nucl. Chem. 30, 2571 (1968).
[12]C. E. Moss, C. Détraz, C. S. Zaidins, and I. J. Frantsvog, Phys. Rev. C 5, 1122 (1972).
[13]A. Gallmann, F. Jundt, E. Aslanides, and D. E. Alburger, Phys. Rev. 179, 921 (1969).
[14]C. E. Moss, C. Détraz, and C. S. Zaidins, Nucl. Phys. A170, 111 (1971).
[15]C. N. Davids, D. L. Matthews, and D. P. Whitmire, Bull. Am. Phys. Soc. 17, 71 (1972); and to be published.
[16]W. D. Arnett and S. E. Woosley, private communication.

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Study of Some ^{20}F Excited States*

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Excited states of ^{20}F which deexcite via γ-ray cascades through the 823-keV state were studied using the ^{18}O(^{3}He, p) reaction at a beam energy of $E_{^3\text{He}} = 2.88$ MeV. Particle γ-ray angular correlations in a standard collinear geometry were obtained using NaI detectors for observing those γ rays which were in coincidence with proton groups representing excited states up to 4.3 MeV excitation. The analysis of these data involved the simultaneous fitting of the correlations of all possible cascading γ rays from the state under study and resulted in the measurement of spins, branching, and multipole mixing ratios for some excited states and their subsequent electromagnetic deexcitations. Coincident γ-ray spectra were also obtained with a Ge(Li) detector which provided additional information regarding the more complex decay schemes. Previously unreported lifetime information for the 1824- and 1972-keV states was obtained using the Doppler-shift-attenuation method and the ^{18}O($t,n\gamma$)^{20}F reaction at a beam energy of $E_t \simeq 2.85$ MeV. These mean lifetimes are ≤ 65 fsec and ≥ 1.1 psec, respectively. The combined data for the 1824-keV state, as well as recently reported two-particle transfer data, suggest that the 823- and 1824-keV states are the $J^\pi = 4^+$ and 5^+ members, respectively, of the ground-state rotational band. Data obtained regarding the 1972-keV state suggest that this state has a $J^\pi = 3^-$ assignment, while those for the 2195-keV state imply a $J^\pi = 3^+$ assignment. The results of this experiment are compared with recently reported shell-model calculations.

I. INTRODUCTION

The nuclear spectroscopy of the excited states of ^{20}F is not well understood despite a large number of completed experimental investigations. This is due in part to the fact that many of the combinations of targets and reactions normally used with conventional standard investigative techniques do not always yield unambiguous spin assignments. One recent study[1] reviews the available spectroscopic information and compares this information with the results of the Oak Ridge shell-model calculations.[2] These shell-model calculations have been very successful in describing nuclear properties of many nuclei in this mass region and should, as a consequence, be just as successful with the nucleus ^{20}F. One of the resulting features of these calculations (independent of the Hamiltonians used) is a set of rotational-band-like states starting with the $J^\pi = 2^+$ ground state. The predicted $J^\pi = 4^+$ state is consistently positioned in the region of excitation equivalent to 1 MeV, while the $J^\pi = 5^+$ state is generally predicted to be at an excitation energy of about 2 MeV. A recent report[3] points out the consistency between experiment and theory if it is assumed that the 656- and 823-keV states are the $J^\pi = 3^+$ and 4^+ members of this rotational band, although the 823-keV state was known to have spin $J = 2$ or 4. In addition to positive-parity states generated by the presence of the active nucleons in sd-shell orbits as calculated by the Oak Ridge shell-model group,[2] it is expected[4] that there should be some low-lying negative-parity states whose origins are outside of the sd shell.

The present investigation consisted of a study of the excited states of ^{20}F using the ^{18}O(^{3}He, $p\gamma$)-^{20}F and ^{18}O($t, n\gamma$)^{20}F reactions at bombarding energies of $E_{^3\text{He}} = 2.88$ and $E_t = 3.0$ MeV. The investigation was undertaken in an effort to better under-