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Radiation from Ga^{64}†

THOMAS H. JACOBI*
Department of Physics, University of California, Los Angeles, California

H. A. HOWE
United States Naval Radiological Defense Laboratory, San Francisco, California

AND

J. REGINALD RICHARDSON
Department of Physics, University of California, Los Angeles, California

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The β^+ decay of Ga^{64} and the subsequent gamma spectrum from the de-excitation of Zn^{64} have been studied. The β^+ transition to the ground state of Zn^{64} has an end point energy of 6.05 ± 0.03 Mev with $\log ft=6.6$ and another β^+ group or groups has an end point energy of 2.79 ± 0.08 Mev with a $\log ft=4.6$. No β^+ transition to the first excited state of $Zn(2+)$ was observed and it was shown that the $\log ft$ corresponding to this transition is greater than 7.7. The gamma rays that were observed (0.80, 0.99, 1.25, 1.38, 1.56, 1.78, 1.95, 2.18, 2.34, 2.99, and 3.32 Mev) indicate excited states at 0.99(2+), 1.78(2+), and 3.32(1+) Mev.

The positron results indicate that the ground state of Ga^{64} has spin and parity 0+.

INTRODUCTION

GALLIUM-64 is a short-lived radioisotope which decays by positron emission to Zn^{64} with a half-life of 2.6 minutes. This decay has been investigated by Cohen,[1] using scintillation techniques, who reported positrons of energies greater than 5 Mev and gamma rays of 1.0 and 3.8 Mev, the latter being questionable. Except for half-life measurements, no other investigations of this decay have been reported. Weller and Grosskreutz,[2] using the reaction $Cu^{63}+p\rightarrow Zn^{64*}$, reported 10 gamma rays and presented a possible energy level diagram and de-excitation scheme. However the scheme is not unambiguous and the only level that seems certain is that at 0.99 Mev which has also been observed by Heydenberg and Temmer[3] using Coulomb excitation and by Van Patter *et al.*[4] by inelastic proton scattering.

THE BETA-RAY SPECTRUM

Gallium-64 was produced through the reaction $Zn^{64}+p\rightarrow Ga^{64}+n$ by bombarding natural zinc foils with protons in the UCLA cyclotron. The positron spectrum was investigated in an iron free thick lens beta spectrometer which was capable of focusing 5.75-Mev positrons. Because of the short half-life of 2.6 min, the sample had to be transported quickly from the cyclotron and positioned within the spectrometer. One point in the beta spectrum was obtained per bombardment by counting for a fixed time with the spectrometer set at the momentum in question and then counting for another fixed time with the spectrometer set at a standard momentum for normalization purposes. Usual subtraction procedures were used for the elimination of activities due to longer-lived radionuclides and each foil was given time for cooling off after each run.

In order to check the linearity of the spectrometer, the end point energy of the Ga^{66} positron spectrum was compared with the conversion electron from Ba^{137} (Table I). The ratio of energies as found from the spectrometer currents was found to agree with the ratio of best values measured by others to within 0.3%. A Kurie plot of an experimental positron spectrum of Ga^{64} over the range of 2.2 to 5.7 Mev is shown in Fig. 1, while Fig. 2 is a Kurie plot of the high-energy end of this spectrum from another run. Each point of Fig. 2 has an error of 1% due to counting statistics. The high-energy beta group end point can be set quite accurately at 6.05 ± 0.03 Mev from this plot. The best value for the energy of the low-energy group is that of 2.79 ± 0.08 Mev found by beta-gamma coincidence as described later. The relative number of decays in the 6.05 and 2.79-Mev groups was calculated as follows.

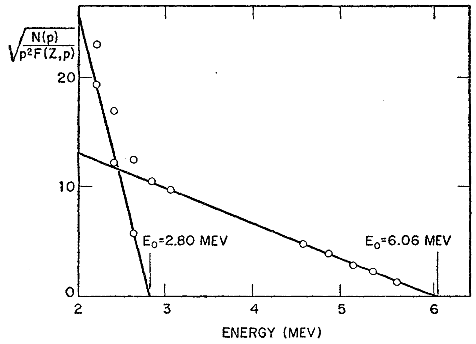

FIG. 1. Kurie plot of the positron spectrum of Ga^{64}.

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* Now at Emerson-Electric of Saint Louis, Saint Louis, Missouri.

[1] B. L. Cohen, Phys. Rev. **91**, 74 (1953).

[2] C. E. Weller and J. C. Grosskreutz, Phys. Rev. **99**, 655(A) (1955).

[3] G. M. Temmer and N. P. Heydenburg, Phys. Rev. **104**, 967 (1956).

[4] D. M. Van Patter, M. A. Rothman, W. C. Porter, and C. E. Mandeville, Phys. Rev. **107**, 171 (1957).

For an allowed decay, with end point W_0, the positron distribution is given by

$$N_w(W_0)=K^2(W_0)F(Z,W)p_w^2(W_0-W)^2, \quad (1)$$

where $K^2(W_0)$ is proportional to the nuclear matrix element of that decay which has an end point of W_0, $F(Z,W)$ is the Fermi factor which is available in tabular form.[5] Integrating this equation for the total number of disintegrations per unit time yields

$$N_T(W_0)=K^2(W_0)f(Z,W_0), \quad (2)$$

where $f(Z,W_0)$ is a function which has been calculated by Feenberg and Trigg.[6]

Since the Kurie plot is a representation of Eq. (1), the quantity $K(W_0)$ is simply minus the slope of the Kurie plot itself and the desired ratio is given by Eq. (3) and found to have the value of 0.33.

$$\frac{N_T(6.05)}{N_T(2.79)}=\frac{K^2(6.05)f(30,6.05)}{K^2(2.79)f(30,2.79)}$$
$$=\left[\frac{\text{Slope of 6.05-Mev Kurie plot}}{\text{Slope of 2.79-Mev Kurie plot}}\right]^2 \times\frac{f(30,6.05)}{f(30,2.79)}. \quad (3)$$

If we use this number for the relative number of disintegrations and a value of 2.6 min for the over-all half-life, the partial half-lives p are

$$T_p(6.05)=624 \text{ sec}, \quad T_p(2.79)=208 \text{ sec}.$$

The corresponding $f(Z,W_0)$ values lead to

$$\log f(30,6.05)=3.8, \quad \log f(30,2.79)=2.3.$$

Therefore the $\log ft$ values are

$$\log ft(6.05)=6.6, \quad \log ft(2.79)=4.6.$$

K capture has been ignored in all these calculations since it would amount to only 3% for an allowed transition with end point 2.79 Mev.

TABLE I. Ga^{64} beta decay.

Beta group end point (Mev)	Abundance	Log*ft*
6.05±0.03	25%	6.6
2.79±0.08	75%	4.6
5.06	0.3±0.4%	7.7

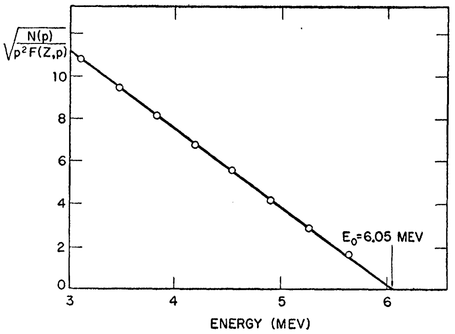

FIG. 2. Kurie plot of the high-energy positron spectrum of Ga^{64}.

BETA-GAMMA COINCIDENCE SPECTRUM

By incorporating a sodium iodide scintillation counter close to the source in the beta spectrometer, beta spectra in coincidence with the various gamma rays of Zn^{64} could be investigated. In order to correct for chance coincidences with this rapidly decaying source, two coincidence systems were used: one for recording real coincidences, and the other, with a delay line to eliminate real counts, for recording accidental coincidences. For any given run the number of true coincidences was given by $N_t=N_r-(\tau_R/\tau_a)N_a$ where N_r and N_a are the number of counts registered in the "real" and "accidental" coincidence circuits and τ_R/τ_a is the ratio of the resolving times of the two circuits. It was also found that over the range of magnetic fields used in the spectrometer the pulse heights from the gamma scintillation counter varied less than 2%. Since all points in the beta spectrum were normalized to the number of gamma-ray counts, this shift had neglible effect.

In particular, it was of interest to use this beta-gamma coincidence technique to eliminate the high-energy beta group and perhaps be able to observe the beta transition from the ground state of Ga^{64} to the 2+ first excited state of Zn^{64} at 0.99 Mev. The Kurie plot of the beta spectrum in coincidence with the 0.99-Mev gamma ray is presented in Fig. 3. All the points have statistical accuracies of 4% and the straight line giving the best fit indicates an end point energy of 2.79±0.08

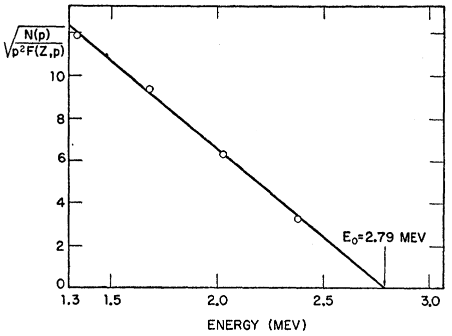

FIG. 3. Beta spectrum in coincidence with 0.99-Mev gamma ray.

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[5] M. E. Rose in *Beta- and Gamma-Ray Spectroscopy*, edited by K. Siegbahn (North Holland Publishing Company, Amsterdam, 1955), p. 875.

[6] See reference 5, p. 285.

Mev. There was no indication of a beta group with a higher end point energy. The lower limit for the *ft* value for the beta transition to the first excited state with end point of 5.06 Mev was estimated in the following way. Many runs were made to determine the number of coincidences between the 0.99-Mev gamma ray and beta particles for two settings of the spectrometer corresponding to 2.20 and 3.55 Mev, normalizing the results as before to the number of gamma rays. Assuming that all coincidences at 2.20 Mev were due to the beta group with end point of 2.79 Mev, and that all coincidences at 3.55 Mev were due to a group going to the first excited state with end point of 5.06 Mev, the following calculations were made. Taking the ratio of the total number of disintegrations, using Eqs. (1) and (2) of the previous section, Eq. (4) is obtained

$$\frac{N_T(5.06)}{N_T(2.79)}=\frac{f(30,5.06)}{f(30,2.79)}\frac{N_{3.55}(5.06)}{N_{2.20}(2.79)}\frac{P_{2.20}^2}{P_{3.55}^2}\times\frac{(2.79-2.20)^2}{(5.06-3.55)^2}\frac{F(30,2.20)}{F(30,3.55)}. \quad (4)$$

Experimentally the ratio $N_{3.55}(5.06)/N_{2.20}(2.79)$ was found to be 0.0075±0.010 which gives for the ratio of the total disintegrations $N_T(5.06)/N_T(2.79)=0.003\pm0.004$. If we choose the upper limit of this ratio, 0.007, the partial half-life for this decay would be $\tau_P(5.06)=6.9\times10^4$ sec and, since $\log f(30,5.06)=3.6$, $\log ft(5.06)=7.7$. Thus experimentally there is no evidence for the occurrence of this transition, but a lower bound on its comparative half-life has been found.

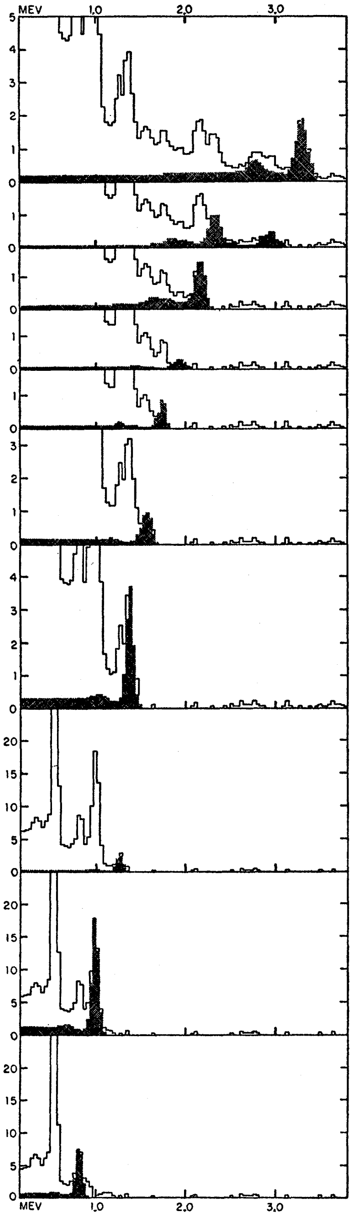

FIG. 4. Analysis of the Zn^{64} gamma spectrum illustrating the method of successive subtraction.

TABLE II. Zn^{64} gamma rays.

Gamma-ray energy (Mev)	Relative intensity
0.35±0.05	...
0.80±0.02	35
0.95±0.05	...
0.99±0.005	100
1.25±0.02	16
1.38±0.02	33
1.56±0.03	16
1.78±0.03	12
1.95±0.04	6
2.18±0.04	25
2.34±0.04	20
2.99±0.05	4
3.32±0.05	41

GAMMA-RAY SPECTRUM

The work on the gamma-ray singles spectrum was done at the U. S. Naval Radiological Defense Laboratory and the Ga^{64} was produced as before from the reaction $Zn^{64}(p,n)Ga^{64}$, using the Crocker cyclotron at the University of California at Berkeley. The spectrometer[7] consisted of a 4×4 inch sodium iodide crystal with the incident radiation almost parallel and collimated to $\frac{1}{2}$ inch diameter. The output from the photomultiplier was amplified and recorded on a 100-channel pulse-height analyzer. Since no high-energy gamma source was available it was decided to use annihilation radiation at 0.51 Mev and the 0.99-Mev gamma ray occurring in this spectrum for calibration. Figure 4 illustrates the method of analysis used on the gamma-ray spectrum of Ga^{64}. In order to obtain the relative intensities of the gamma rays the calibration results obtained by USNRDL personnel for this crystal and collimator geometry were used. In particular the response or profile of the spectrometer has been measured

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[7] C. S. Cook and F. M. Tomnovec, Phys. Rev. **104**, 1407 (1956).

for many gamma rays with energies up to 2.75 Mev. By starting at the high-energy end of the Zn^{64} gamma spectrum, subtracting off the corresponding profile, taking each gamma ray in sequence and interpolating between the standard profiles when necessary, it is possible to determine accurately the number of pulses in the full energy peak of each gamma ray of the spectrum. It is estimated that these numbers are accurate to better than 5% of the most intense gamma-ray round. Once the relative number of pulses in the full energy peak has been found, the following three factors must be used to determine the relative intensities of the gamma rays: (1) the ratio of the total number of pulses corresponding to a given gamma ray to the number of pulses in the full energy peak,[8] (2) the probability that a gamma ray, reaching the face of the sodium iodide crystal, will be converted in any way in the crystal,[9] and (3) the relative aperture of the collimating system which is a function of the gamma-ray penetration and hence of the gamma-ray energy.[10,11] The relative intensities in Table II were obtained after the relative number of pulses in the full energy peaks were altered by these three factors.

TABLE III. Zn^{64} gamma-gamma coincidences.

Gamma rays "looked at" (Mev)	Gamma rays in coincidence
0.80	0.99, 1.24, 1.38, 1.56 Mev
0.99	0.80, 1.0, 1.25, 1.38, 1.95, 2.18, 2.34
1.25	0.80, 0.99, nearly equal Nos.
1.38	0.80, 0.99, more of the 0.99

GAMMA-GAMMA COINCIDENCE SPECTRA

In order to define more completely the decay scheme, gamma-gamma coincidence measurements were made at UCLA. Two sodium iodide scintillation counters were used, the output of one going to a differential pulse-height analyzer, and the output of the other activating the vertical deflection of an oscilloscope. With the discriminator set on the full energy peak of a particular gamma ray, the oscilloscope, registering pulses from the other counter, was triggered only when a coincidence occurred between the two counters. The traces on the oscilloscope were photographed and the results of twelve such photographs are summarized in Table III.

SUMMARY OF RESULTS AND DISCUSSION

It will be assumed that the high-energy beta group corresponded to a transition to the ground state of Zn^{64}, since a recent determination[12] of the $Zn^{64}(p,n)Ga^{64}$ reaction threshold indicates a maximum beta energy for the inverse transition to the ground state of Zn^{64} of 6.0 ± 0.15 Mev. Assuming there are no low lying excited states in Zn^{64}, the high-energy beta group must correspond to a transition to the ground state of Zn^{64}.

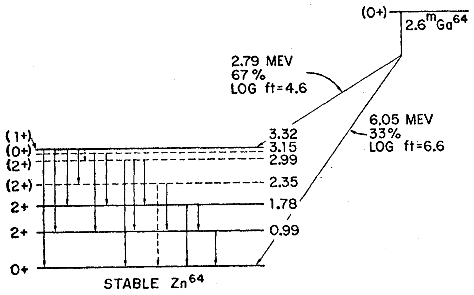

FIG. 5. Proposed decay scheme of Ga^{64}.

This positron decay between ground states has a $\log ft$ value of 6.6. Since the ground state of Zn^{64} has spin and parity of 0+, and Ga^{64} is expected to have + parity, this is an allowed decay but rather unfavored. Since only + parities are involved in the transition to the first excited state, it must either be allowed or second forbidden. With a $\log ft \geq 7.7$ an allowed transition would be unfavored by a factor of 10^4 which does not seem likely, and therefore this transition is classified as second forbidden. Therefore, with the assumption that the parity of the ground state of Ga^{64} is + and the classification of the transitions to the ground state and to the first excited state of Zn^{64} as sllowed and second forbidden, respectively, the ground state of Ga^{64} must be 0+. Since Ga^{64} is an odd-odd nucleus with both unpaired particles in either $f_{\frac{5}{2}}$ or $p_{\frac{3}{2}}$ states, Nordheim's coupling rules would predict a spin greater than 0. These rules are contradicted not only by this experiment indicating 0 spin for the ground state of Ga^{64} but also by the direct measurement[13] of zero spin for the ground state of Ga^{66}.

The lower energy beta group has a $\log ft$ of 4.6, and hence is a normal allowed transition or a combination of several allowed transitions of nearly equal energy. The gamma-ray measurements indicate that the latter statement is more probable, since the most consistent explanation of gamma energies, intensities, and coincidences has levels at 2.99, 3.15, and 3.32 Mev. Figure 5 shows this scheme with the less certain levels shown by dashed lines. The reasoning behind this assignment of levels is as follows:

(a) The 3.32, 2.99, and 1.78-Mev gamma rays do not coincide with the 0.99-Mev gamma ray, and hence represent transitions to the ground state from higher levels.

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[8] F. M. Tomnovec (private communication).

[9] W. F. Miller, J. Reynolds, and W. J. Snow, Rev. Sci. Instr. **28**, 717 (1957).

[10] R. L. Mather, J. Appl. Phys. **28**, 1200 (1957).

[11] C. S. Cook, Bull. Am. Phys. Soc. **1**, 378 (1956).

[12] H. A. Howe, Phys. Rev. **109**, 6 (1958).

[13] J. C. Hubbs, W. A. Nierenberg, and H. A. Shugard, Phys. Rev. **105**, 1928 (1957).

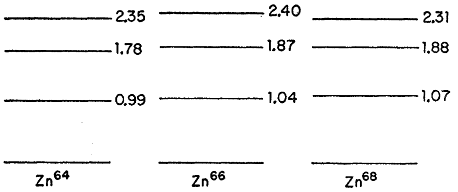

FIG. 6. Comparison of excited states of Zn^{64}, Zn^{66}, and Zn^{68}.

(b) The 2.18-Mev gamma-ray coincides with the 0.99-Mev gamma ray but not with the 0.80-Mev gamma ray. It seems most probable that it is a transition from a level at 3.17 to that at 0.99 Mev.

(c) After the above considerations there remains one ambiguity, the occurrence of the 1.0-Mev gamma ray in coincidence with the 0.99-Mev gamma ray. This can be explained by the existence of a level at about 2.0 or 2.35 Mev. Of these two possibilities the latter is preferable for the following reason. Recent scattering experiments by Cohen[14] indicate that there is no Zn level at about 2.0 Mev. His results with natural Zn targets show strong peaks at 1.57 and 2.30 Mev and no indication of a peak between.

The tentative spin assignments of the excited states of Zn^{64} are in good agreement with the relative gamma-ray intensities and are primarily determined by the ratio of transition probability to the first excited state, 2+, and the ground-state 0+. The 1+ assignment to the level at 3.32 Mev is felt to be considerably more certain than the other assignments because it is supported by both the beta- and gamma-ray information. The level at 1.78 Mev is believed to be 2+ since a higher spin would not be expected to give a measurable amount of crossover radiation, while a lower spin would cause an allowed or first forbidden beta transition to this level, which was not observed. The best data available on the first three excited states of the isotope Zn^{64}, Zn^{66}, and Zn^{68} are compared in Fig. 6. Here, with increasing number of neutrons one sees a slow increase in energy for the first two excited states.

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[14] B. L. Cohen and A. G. Rubin, Phys. Rev. **111**, 1568 (1958).