Disintegration of Ga^{70}†

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The radiations of Ga^{70} (21.1 min) have been investigated with beta- and gamma-scintillation spectrometers. The principal mode of decay is a ground-state beta transition of energy 1.65 ± 0.01 Mev. Study of the gamma spectrum has revealed two gamma rays with energies and intensities (relative to the total number of disintegrations) of 1.036 ± 0.010 Mev (0.76%) and 0.174 ± 0.005 Mev (0.44%). These two gamma rays are in coincidence. From the above facts and the results of beta-gamma coincidence studies, it is concluded that there are two weak beta groups with energies and intensities of 0.61 ± 0.02 Mev (0.3%) and 0.44 ± 0.02 Mev (0.5%) leading from Ga^{70} to levels in Ge^{70} at 1.036 and 1.21 Mev, respectively. The angular correlation of the two gamma rays has been measured, and the observed correlation function is in agreement with that expected for the spin sequence 0–2–0. A search for evidence of the 1.21-Mev electric-monopole crossover transition yielded negative results.

INTRODUCTION

GALLIUM-70 (21 min) decays to the even-even nucleus Ge^{70} by beta emission. The most extensive previous study of this isotope was made by Haynes,[1] who examined the beta spectrum with a magnetic-lens spectrometer. The associated Kurie plot had an end point of 1.65 Mev and was linear down to about 0.4 Mev. It was not determined whether the deviation from a straight line below 0.4 Mev resulted from scattering or another beta group. Except for the observation that there were no apparent conversion lines above 0.6 Mev, no gamma-ray information was obtained.

Prior to the present investigation, it had been reported[2] that gamma rays of energy 1.04 and 2.0 Mev are associated with the decay of As^{70} to Ge^{70}. On the basis of a survey of first excited states of even-even nuclei,[3,4] Scharff-Goldhaber and McKeown[4] proposed that the 1.04-Mev gamma ray is a transition from the first excited state of Ge^{70} to the ground state. Since it seemed probable that the spin of Ga^{70} is 1+ and the spin of the first excited state of Ge^{70} is 2+, there was reason to suspect that Ga^{70} should exhibit detectable beta branching to the hypothetical 1.04-Mev level of Ge^{70}. This hypothesis prompted the present reinvestigation of the radiations of Ga^{70}.

SOURCE PREPARATION

The Ga^{70} sources were prepared by neutron irradiation of gallium enriched in Ga^{69} (98.42 percent Ga^{69}, 1.58 percent Ga^{71}). The enriched gallium, obtained from the Stable Isotopes Division of Oak Ridge National Laboratory, was in the chemical form Ga_2O_3. Since the spectrographic analysis indicated that none of the detectable impurities had an abundance of $>0.08\%$, no further chemical purification of the irradiated gallium was made. However, the irradiated samples did contain the radioactive "contaminant" Ga^{72} (14-hr). Therefore, all Ga^{70} spectra shown in this paper were obtained by first measuring the composite ($Ga^{70}+Ga^{72}$) spectrum, later determining the Ga^{72} contribution, and then appropriately subtracting the Ga^{72} spectrum from the gross data.

HALF-LIFE MEASUREMENTS

To facilitate accurate time correction of the observed data, the Ga^{70} and Ga^{72} half-lives were measured both with an end-window methane flow counter and with a large 4π proportional counter. The ratio of Ga^{70} to Ga^{72} beta activity was initially $\sim$500:1. From a semi-logarithmic plot of the data, the Ga^{72} half-life was estimated to be 14.2 hr, in good agreement with previous determinations. A function of the form $N=N_0e^{-\lambda t}$ was then fitted to the net Ga^{70} data by a least-squares procedure adapted to IBM machine calculation. Only the data taken during the first hour of decay were analyzed. The average of the half-life values obtained for Ga^{70} was found to be 21.10 ± 0.07 min.[5]

GAMMA-RAY SPECTRUM

The gamma-ray spectrum of Ga^{70}, shown in Fig. 1, was obtained with a 2×2-inch NaI(Tl) crystal mounted on a Dumont 6292 photomultiplier. A ten-channel analyzer was used for the recording of data. Measurements were begun a few minutes after bombardment, at which time approximately half of the gross counting rate resulted from Ga^{72} gamma activity. The net spectrum shown in Fig. 1 was found to decay with the half-life of Ga^{70}. There are two well-resolved photopeaks with energies of 0.174 ± 0.005 and 1.036 ± 0.010 Mev. The gamma rays of Ce^{139} (166.5 kev) and Na^{22} (1.275 Mev) were used for energy calibration. The fast rise of the spectrum at low energy is attributed to

† Work performed under the auspices of the U. S. Atomic Energy Commission.
[1] S. K. Haynes, Phys. Rev. **74**, 423 (1948).
[2] A. H. Wapstra and N. F. Verster, cited by B. Verkerk and A. H. W. Aten, Jr., Physica **18**, 974 (1952).
[3] G. Scharff-Goldhaber, Phys. Rev. **90**, 587 (1953).
[4] G. Scharff-Goldhaber and M. McKeown, Phys. Rev. **92**, 356 (1953).

[5] An error was found in the calculations which yielded the value of 21.37 min reported previously: Bunker, Mize, and Starner, Phys. Rev. **95**, 612(A) (1954).

bremsstrahlung associated with the 1.65-Mev beta group.

The absolute intensities (per beta disintegration) of the 0.174- and 1.036-Mev gamma rays were determined in the following manner. First, two Ga^{70} sources were prepared which had relative activities of 1210:1, as measured with a Geiger counter arrangement involving two source-counter geometries with known relative counting efficiencies for the P^{32} beta activity (end-point energy: 1.71-Mev). The gamma spectrum of the stronger source was then observed with a NaI(Tl) crystal whose absolute "photopeak efficiency" as a function of energy was known for the source-detector geometry used. The weaker sample, which had an average source-plus-backing thickness of <0.1 mg/cm^2, was transferred to a 4π proportional counter with which the absolute beta disintegration rate was determined. From these data, absolute intensities of 0.44 ± 0.09 and $0.76\pm0.08\%$, respectively, were obtained for the 0.174- and 1.036-Mev gamma rays.

To investigate the possibility that the two observed gamma rays were in cascade, we performed a gamma-gamma coincidence experiment[6] in which pulses in the 1.036-Mev photopeak were used to "gate" the ten-channel analyzer. Possible coincidences resulting from Compton scattering of the 1.036-Mev quanta were eliminated by orienting the scintillators so that they subtended an angle of 90° at the source and then shielding the detectors from one another with 1/4-inch of lead. A strong peak was observed in the coincidence spectrum at 174 kev, only a few percent of which resulted from chance events. This peak was found to decay with a 21-min half-life. The two gammas are therefore definitely in cascade. No other photopeaks were found in the coincidence spectrum.

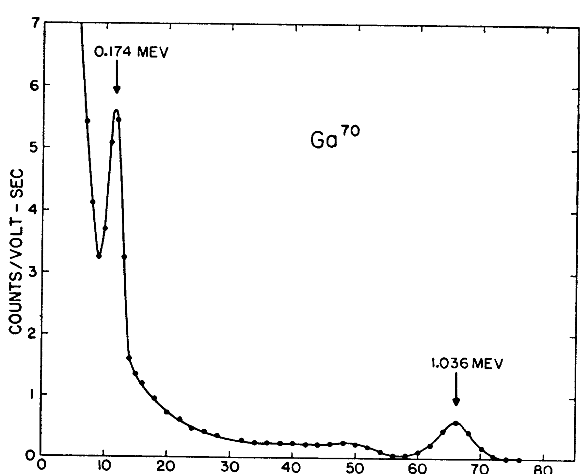

FIG. 1. Scintillation spectrum of Ga^{70}, obtained with a 2×2-inch NaI(Tl) crystal.

[6] For a description of the coincidence apparatus, see Bunker, Mize, and Starner, Phys. Rev. 94, 1694 (1954).

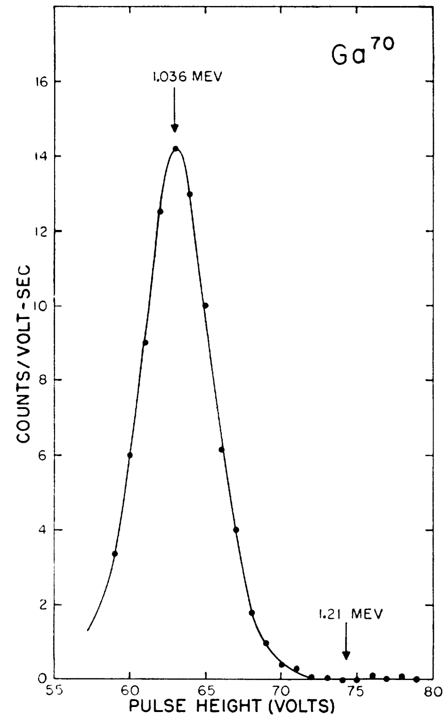

FIG. 2. High-energy portion of Ga^{70} scintillation spectrum.

A careful search was made for evidence of the 1.21-Mev crossover gamma transition, with the negative result shown in Fig. 2. On the basis of these data, it is estimated that the intensity of the hypothetical crossover gamma transition is $\leq 0.5\%$ of the intensity of the 1.036-Mev transition.

BETA-RAY MEASUREMENTS

The end-point energy of the ground-state beta transition was remeasured with a magnetic-lens spectrometer. A value of 1.65 ± 0.01 Mev was obtained, in agreement with the measurement of Haynes.[1]

No attempt was made to obtain the end-point energies of the lower-energy groups from the lens spectrometer data since it was known from the absolute gamma intensity measurements that the sum of the intensities of these groups was $<1\%$. Instead, the lower-energy beta groups were observed as beta-gamma coincidence spectra. A Pilot Plastic Scintillator-B[7] phosphor, $1\frac{3}{4}$ inches in diameter and 3/4 inch thick, served as the beta detector, and a 2×2-inch NaI(Tl) crystal was used as the gamma detector. The beta spectrum in coincidence with the 1.036-Mev photopeak was found to have an end-point energy of ~ 0.6 Mev, and the beta spectrum in coincidence with the 0.174-Mev photopeak was found to have an end-point energy of ~ 0.4 Mev. Thus the 1.036-Mev gamma is a ground-state transition and is preceded by the 0.174-Mev

[7] Pilot Chemicals, Inc., 47 Felton Street, Waltham 54, Massachusetts.

transition. Furthermore, these experiments prove that there are direct beta transitions to levels of Ge^{70} at 1.036 Mev and 1.21 Mev. From the energies of the two gammas and the energy of the ground-state beta transition, the end-point energies of the two low-energy beta groups are calculated to be 0.61±0.02 and 0.44±0.02 Mev.

ANGULAR CORRELATION MEASUREMENT

The log*ft* values of the observed beta groups indicated that all three transitions were undoubtedly of the allowed type, which implied that the two excited states must have spins of 0, 1, or 2 and even parity. Since the first excited state of an even-even nucleus is usually a 2+ state,[3] and since in the present case no crossover transition from the second excited state to the ground state was observed, the most plausible guess for the spin sequence of the three "observed" levels of Ge^{70} was 0–2–0. The fact that the 0–2–0 angular correlation function exhibits such an extreme degree of anisotropy makes it easy to distinguish from any pure or mixed correlation of the types 1–2–0 or 2–2–0. Therefore, it was believed that an angular correlation experiment might be informative in spite of the relatively poor data which one could expect because of experimental difficulties associated with the short half-life.

The gamma-gamma directional correlation pattern was studied by measuring the coincidence counting rate as a function of the angle subtended at the source by two NaI(Tl) detectors. Both NaI(Tl) crystals were 2×2-inch cylinders. The crystals were shielded from direct beta rays with 3/8-inch thick Lucite (see Fig. 3). The half-width at half-maximum of the angular resolution curve was measured to be 11.0°.

Conventional electronic circuits were used. The resolving time of the coincidence circuit was $2\tau \simeq 4.0 \times 10^{-7}$ sec. Both channels of the coincidence circuit were set to accept all pulses corresponding to energies greater than 145 kev.

A typical run proceeded as follows: a 5-mg sample of enriched Ga_2O_3 was irradiated for 30 minutes in a neutron flux of $\sim 6\times 10^{11}$ neutrons/cm²-sec. The radioactive powder was dissolved in 50λ of hot HCl and then transferred to a Lucite source holder of the type shown in Fig. 3. Data were taken at only one angle in a given run. Data-taking began about 7 minutes after the end of the bombardment and continued for 30 to 45 minutes. Several hours later, the 14-hr Ga^{72} background was measured. About half of the total observed coincidences were found to be true Ga^{70} gamma-gamma coincidences; the rest were due to Ga^{72}, cosmic background, chance events, and Compton backscattering of the 1.04-Mev Ga^{70} gamma ray. The magnitude of the Compton backscattering correction was determined by measuring the coincidence rate resulting from scattering of the 1.08-Mev gamma ray of Rb^{86}. This correction amounted to about 5% at 169°. The net Ga^{70} gamma-gamma coincidence rate was normalized by dividing by the total Ga^{70} contribution to the singles rate in the fixed counter.

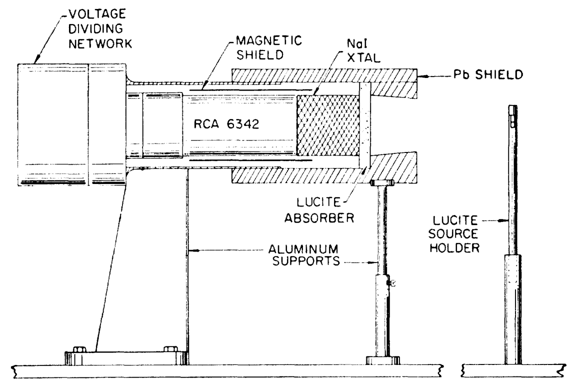

FIG. 3. Scintillation counter arrangement used in the angular correlation measurement.

TABLE I. Log*ft* values of the beta groups of Ga^{70}.

Transition energy (Mev)	Percent branch[a]	Log*ft*[b]
1.65±0.01	99.2 ±0.2	5.10
0.61±0.02	0.28±0.13	5.95
0.44±0.02	0.48±0.10	5.16

[a] Calculated from the observed absolute gamma intensities and the theoretical internal conversion coefficients [Rose, Goertzel, and Perry, Oak Ridge National Laboratory Report ORNL-1023, 1951 (unpublished); Rose, Goertzel, and Swift (privately circulated tables)]. It was assumed that the 0.174- and 1.036-Mev transitions are pure *E*2 (see text).

[b] Determined from the curves given by S. A. Moszkowski, Phys. Rev. **82**, 35 (1951).

Observations were made at four settings of the variable angle θ: 90°, 130°, 150°, and 169°. The experimental results, corrected for the finite geometry, are shown in Fig. 4. The solid curve is the theoretical curve for the 0–2–0 correlation, and the dashed curve is the least-squares fit of the experimental points to a curve of the form $W(\theta)=1+A_2P_2(\cos\theta)+A_4P_4(\cos\theta)$. The experimental coefficients are $A_2=0.444\pm 0.128$ and $A_4=1.303\pm 0.212$. Since the theoretical coefficients for the 0–2–0 correlation ($A_2=0.357$, $A_4=1.143$) fall inside the quoted errors in the experimental coefficients and none of the 1–2–0 or 2–2–0 pure or mixed correlations has a value for A_4 exceeding +0.327, we conclude that the angular correlation data definitely establish the spin sequence as 0–2–0.

FURTHER STUDY OF THE DECAY CHARACTERISTICS OF THE 1.21-MEV LEVEL

Since the lifetime (τ_γ) of the 0.174-Mev *E*2 transition calculated on the basis of the Weisskopf single-particle model[8] is $\sim 3\times 10^{-7}$ sec, an attempt was made to measure the half-life of the 1.21-Mev level. A conventional fast-slow delayed-coincidence scintillation spectrometer with a resolving time of $2\tau \simeq 2\times 10^{-8}$ sec was used for this measurement. The experiment consisted of measuring the time distribution of coincidences between pulses in the 125- to 400-kev portion of the Ga^{70}

[8] V. F. Weisskopf, Phys. Rev. **83**, 1073 (1951).

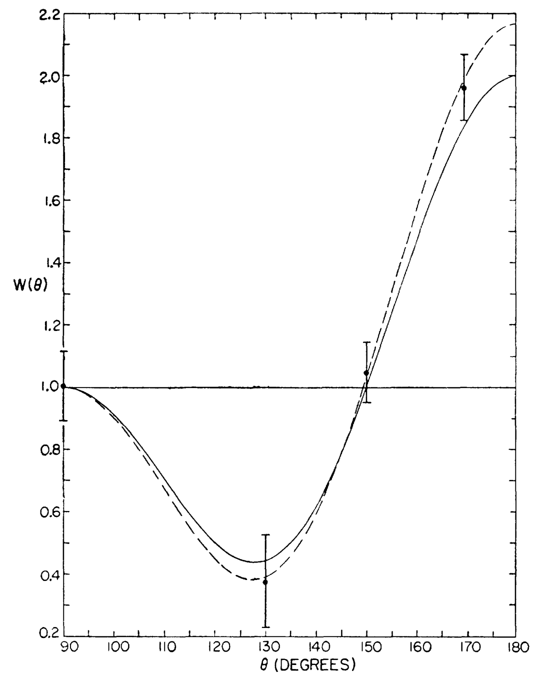

FIG. 4. Angular correlation results. The dashed curve represents the least squares fit to the indicated experimental points. The solid curve is the theoretical 0–2–0 correlation.

beta spectrum (observed with a Pilot[7] plastic scintillator) and pulses in the 0.174-Mev photopeak [observed with a NaI(Tl) crystal]. Proof that the observed coincidences resulted almost entirely from (0.44-Mev β, 0.174-Mev γ) events was established by analyzing the coincident spectra with gated multichannel analyzers. The delayed-coincidence data indicate that the half-life of the 1.21-Mev level is $\leq 4\times 10^{-9}$ sec.

It was also of interest to look for evidence of 1.21-Mev electric-monopole transitions to the ground state. Such a transition can proceed only by pair emission or internal conversion. A careful search for pair emission was made by the standard technique of looking for coincidences between annihilation quanta. The two gamma detectors, both 2×2-inch NaI(Tl) crystals, were positioned in a 180° geometry with the source midway between them. The Lucite source-holder walls were thick enough to stop 600-kev positrons. From a comparison of the (0.511-, 0.511-Mev) coincidence counting rates associated with sources of Ga^{70} and Na^{22}, whose relative disintegration rates were determined by comparing the intensities of the Ga^{70} 1.04-Mev and Na^{22} 1.275-Mev gamma rays, an upper limit of 2×10^{-4} can be placed on the ratio of the transition probability of 1.21-Mev monopole pair emission to that of 0.174-Mev gamma emission.

The search for evidence of 1.2-Mev conversion electrons was carried out by means of a similar coincidence technique. The (0.44-Mev β, 1.2-Mev e^-) coincidence rate per 0.44-Mev beta of a Ga^{70} source was compared with the (0.97-Mev β, 0.33-Mev e^-) coincidence rate per 0.97-Mev beta of a Au^{198} source. In the Ga^{70} coincidence measurement, the beta channel was set to accept pulses corresponding to the energy range 100 to 400 kev. The fraction of these pulses which was associated with the 0.44-Mev beta group was estimated from the measured beta-branching ratios and the empirically-determined spectral shapes of the three beta groups. From the above comparison measurement and the known K-conversion coefficient[9] of the 0.412-Mev Hg^{198} gamma transition, an upper limit of 4×10^{-2} was calculated for the ratio of the transition probability of 1.21-Mev monopole internal conversion to that of 0.174-Mev gamma emission.

The above results indicate that the lifetime of the 0.174-Mev gamma transition is essentially the same as the lifetime of the 1.21-Mev level, i.e., $(\tau_\gamma)_{0.174}\leq 6\times 10^{-9}$ sec. Although this value is ≥ 50 times smaller than the Weisskopf estimate, it should be noted that the known $E2$ transition probabilities in the $Z=22$ to $Z=48$ region, determined from Coulomb excitation experiments,[10] are larger than the single-particle estimates by factors ranging from 10 to 60. In fact, the Coulomb excitation results on Ge^{70} indicate that the half-life of the 1.036-Mev ground-state transition is 1.4×10^{-12} sec,[11] which is about a factor of 10 shorter than the single-particle estimate.

Church and Weneser[12] have calculated the transition probability for electric-monopole conversion in the K shell in terms of a nuclear strength parameter, ρ. Their results indicate a partial half-life $(T_{1/2})_K \sim 1.6\times 10^{-7}$ sec for this mode of decay of the 1.21-Mev level if one assumes the same value for ρ as is associated with the 0.69-Mev monopole transition in Ge^{72}.[13] This estimate of $(T_{1/2})_K$, coupled with the measured upper limit on the half-life of the 1.21-Mev level and corrected for the ratio of total internal-conversion probability to K-conversion probability (assumed to be 1.2), yields a value of ≤ 0.03 for the fractional decay probability associated with the monopole-conversion mode of decay. Our corresponding experimental value (≤ 0.04) is compatible with this estimate.

By using Thomas'[14] expression for the monopole internal pair production probability (W_π) and Church and Weneser's[12] expression for the monopole K-conversion probability (W_K), one can arrive at a theoretical estimate for the ratio W_π/W_K. For the 1.21-Mev Ge^{70}

[9] L. Simons, Phys. Rev. **86**, 570 (1952).
[10] G. M. Temmer and N. P. Heydenburg, Phys. Rev. **99**, 1609 (1955).
[11] N. P. Heydenburg and G. M. Temmer, reported in *Nuclear Level Schemes*, $A=40-A=92$, compiled by Way, King, McGinnis, and van Lieshout, Atomic Energy Commission Report TID-5300 (U. S. Government Printing Office, Washington, D. C., 1955), p. 107.
[12] E. L. Church and J. Weneser, Phys. Rev. **100**, 943 (1955); **103**, 1035 (1956).
[13] Kraushaar, Brun, and Meyerhof, Phys. Rev. **101**, 139 (1956).
[14] R. Thomas, Phys. Rev. **58**, 714 (1940).

transition, $W_\pi/W_K \sim 10^{-2}$.[15] If $(T_{1/2})_K$ is assumed to be $\sim 1.6 \times 10^{-7}$ sec, as indicated above, then $(T_{1/2})_\pi \sim 1.6 \times 10^{-5}$ sec. This result, coupled with the measured upper limit on the half-life of the 1.21-Mev level, yields a value of $\leq 2.5 \times 10^{-4}$ for the fractional decay probability associated with the monopole-pair-production mode of decay. The corresponding experimental value of $\leq 2 \times 10^{-4}$ is consistent with this estimate.

DISCUSSION

The decay scheme suggested by the measurements described above is shown in Fig. 5. The spin and parity assignments which have been made are the only ones consistent with the experimental data. Confirmation of the 2+ assignment for the 1.036±0.010-Mev level is given by the fact that a level in Ge^{70} at 1.020 ±0.015 Mev has been observed by Coulomb excitation.[16] Subsequent to these experiments, gamma rays with energies of 0.18 and 1.06 Mev were reported to accompany the decay of As^{70},[17] and although no decay scheme has yet been proposed for As^{70}, it seems probable that these two gammas correspond to the 0.174- and 1.036-Mev transitions of the present experiment.

The 1+ assignment for Ga^{70} is readily explainable on the shell-model basis as resulting from the coupling of a $(p_{3/2})^3$ proton configuration and a $(p_{1/2})^1$ neutron configuration.

The occurrence of a low-lying 0+ level in Ge^{70} may be associated with the possible closed subshell character of the ground state: specifically, a $(p_{3/2})^4$ proton configuration and a $(p_{3/2})^4(f_{5/2})^6$ neutron configuration. Such an explanation has been proposed for the existence of the known 0+ excited levels of Ge^{72},[3] Ca^{40},[18] and Zr^{90}.[19] The assumption of a closed subshell structure for stable Ge^{70} implies that the 1.036-Mev 2+ level and the 1.21-Mev 0+ level involve configurations different from that of the ground state, which provides a possible explanation for the relatively high logft value of the 0.61-Mev beta transition. The fact that the transition

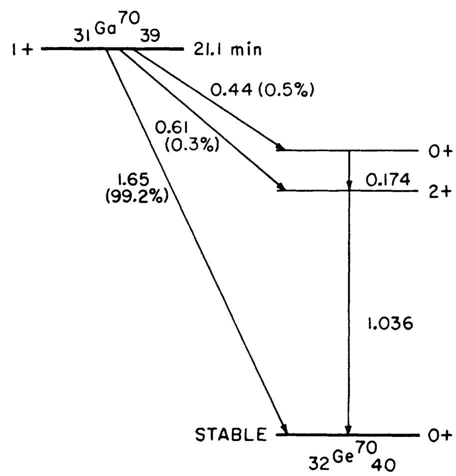

FIG. 5. Proposed decay scheme of Ga^{70}.

probability of the 0.44-Mev beta transition is not similarly reduced can be explained in terms of configuration mixing.[20] It is reasonable that mixed configurations should occur in this region since the $p_{3/2}$ and $f_{5/2}$ states seem to be nearly degenerate.

It can be calculated from the threshold of the $Zn^{70}(p,n)Ga^{70}$ reaction[21] that the Ga^{70} ground state is 0.67 Mev above the ground state of Zn^{70}. The fact that no evidence for K capture was found in the present experiments can be easily understood, however. If the above decay energy is correct, the intensity of the allowed K capture transition to the Zn^{70} ground state would amount to $\leq 1\%$ of the total Ga^{70} disintegration rate and would be very difficult to detect. Decay branches to excited states would be expected to have much smaller intensities, and if the first excited state of Zn^{70} is at ~ 0.7 Mev, as suggested by the existing empirical data,[3,4] then decay to this level may even be energetically impossible.

ACKNOWLEDGMENT

The authors are indebted to Dr. G. Scharff-Goldhaber for suggesting this problem and for helping to plan some of the initial experiments.

[15] The authors are indebted to Dr. Eugene Church for making this calculation.
[16] N. P. Heydenburg and G. M. Temmer, Phys. Rev. **99**, 617(A) (1955); verbal report.
[17] F. D. S. Butement and E. G. Prout, Phil. Mag. **46**, 357 (1955).
[18] Bent, Bonner, and McCrary, Phys. Rev. **98**, 1325 (1955).
[19] K. W. Ford, Phys. Rev. **98**, 1516 (1955).
[20] A detailed discussion of the effects of configuration mixing has been given by A. de-Shalit and M. Goldhaber, Phys. Rev. **92**, 1211 (1953).
[21] C. C. Trail and C. H. Johnson, Phys. Rev. **91**, 474 (1955).