$(96.5\pm0.5)\%M1$ and $(3.5\pm0.5)\%E2$ with $\delta_{82}=-0.190\pm0.014$. The 301-kev gamma ray can have one of the two possible mixtures: either $\delta_{301}=+0.123\pm0.004$ with a mixture of $(98.5\pm1.0)\%M1$, and $(1.5\pm1.0)\%E2$, or $\delta_{301}=-3.98\pm1.02$ with a mixture of $(6\pm2)\%M1$ and $(94\pm2)\%E2$. The value of $\delta_{301}=-3.98$ is more probable. The 80-kev gamma ray is also found to have two possible values of δ_{80}. Either $\delta_{80}=+0.47\pm0.09$ with a mixture of $(82\pm6)\%M1$ and $(18\pm6)\%E2$, or $\delta_{80}=+7.0$ with a mixture of $(2.0\pm1.5)\%M1$ and $(98.0\pm1.5)\%E2$.

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Decay of K^{42} and Sc^{44}†

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The low excited states of Ca^{42} and Ca^{44} have been studied in the decay of K^{42} and Sc^{44}, with special emphasis on the observation of weakly populated states. In the K^{42} decay, gamma rays have been seen at 0.31 Mev (1.1%), 0.49 Mev (<0.1%), 0.60 Mev (0.1%), 0.90 Mev (0.1%), 1.02 Mev (0.1%), 1.52 Mev (100%), 1.92 Mev (0.3%), and 2.42 Mev (0.2%). The coincidence sequence of the transitions has been measured and a level scheme constructed. In the Sc^{44} decay, gamma rays have been seen with energies and intensities of 0.68 Mev (3.2%), 1.02 Mev (3.1%), 1.12 Mev (4.7%), 1.16 Mev (100%), 1.50 Mev (1.7%), 1.72 Mev (0.8%), 2.28 Mev (0.2%), and 2.69 Mev (0.2%). Coincidence measurements were also taken for this isotope to clarify the cascade sequences, and a level scheme was constructed. A search was made for low-energy conversion electrons ($E<1$ Mev) in an effort to establish the existence or nonexistence of a low-lying 0^+ state in Ca^{44}, whose analog occurs as the second excited 1.84-Mev state in Ca^{42}. No such conversion electrons were seen, by either electron spectrometer studies or by electron-delayed gamma-ray coincidence measurements. An upper limit of 0.05% of the total decay of Sc^{44} was put on the population of such a state.

I. INTRODUCTION

NUCLEI that are within a few nucleons of being doubly magic are of special interest from a theoretical point of view. The interest stems from the ability of shell-model calculations to predict certain features of these nuclei in a straightforward manner. The Ca isotopes are of particular interest in this regard, since in the basic shell model they involve the coupling of the $f_{7/2}$ neutrons (between 20 and 28) with themselves, the 20 protons being a stable configuration. Several recent theoretical analyses[1–5] have had to rely on inadequate experimental information concerning the excited states of the Ca isotopes. The present effort is an attempt to obtain more information on the levels of Ca^{42} and Ca^{44} through a study of the decays of K^{42} and Sc^{44}. Although completely quantitative work was made difficult because the transitions of interest were extremely weak, enough new information has been obtained on a number of levels of Ca^{42} and Ca^{44} to clarify spin assignments, and in some cases to compare cascade-to-crossover transition probabilities.

The general features of the K^{42} decay have been summarized by Way *et al.*[6] The dominant decays are a 3.55-Mev beta transition (82%) to the ground state of Ca^{42}, and a 1.99-Mev transition to the first excited state of Ca^{42}. The 3.55-Mev beta-ray spectrum shape has been shown to be consistent with a unique first forbidden transition, and the log *ft* values of both decays indicate first forbiddenness. At the time this experiment was begun, only two gamma-ray transitions were known: the 1.52-Mev transition (100%) from the first excited state, and the 0.31-Mev line (~1%) from the second to the first excited state. During the course of this work two angular correlation measurements have been reported[7,8] for the 0.31–1.52-Mev gamma-ray cascade, and conversion electrons from the 1.84-Mev transition have been seen.[9] Both measurements very clearly indicate a 0-2-0 spin sequence. Three additional

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[1] C. Levinson and K. W. Ford, Phys. Rev. **100**, 13 (1955).
[2] M. K. Banerjee and B. Dutta Roy, Ann. Phys. **7**, 484 (1959).
[3] J. B. French and B. J. Raz, Phys. Rev. **104**, 1411 (1956).
[4] I. Talmi, Phys. Rev. **107**, 326 (1957).
[5] H. E. Mitler, thesis, Princeton University, Princeton, 1960 (unpublished).
[6] K. Way, R. W. King, C. L. McGinnis, and R. van Lieshout, *Nuclear Level Schemes A*=40−*A*=90, Atomic Energy Commission Report TID-5300 (U. S. Government Printing Office, Washington, D. C., 1955).
[7] H. Morinaga, N. Mutsuro, and M. Sugawara, Phys. Rev. **114**, 1146 (1959).
[8] I. Asplund and T. Wiedling, Phys. Rev. **116**, 741 (1959).
[9] N. Benczer-Koller, M. Nessin, and T. H. Kruse, Bull. Am. Phys. Soc. **5**, 248 (1960).

weak transitions have been reported by Morinaga, Mutsuro, and Sugawara[7] at 0.9 Mev (0.3%), 1.94 Mev (0.3%), and 2.42 Mev (0.2%). The observation of these three weak gamma rays paralleled the intent of the present studies and indicated that a further search for additional weak transitions would be fruitful.

In order to study the levels of Ca^{44}, the Sc^{44*} 2.44 day isomer was chosen over K^{44} (22.4 min half-life) for convenience, although the energetics of the decay are not as favorable. The Sc^{44*} isomer proceeds[6] mainly by a 0.27-Mev gamma-ray transition to the ground state of Sc^{44} and then via a 1.46-Mev positron branch to the 1.16-Mev state of Ca^{44}. Other than the 1.16-Mev gamma ray the only other reported[10] has been a line at 2.54 Mev. Information from reaction studies[11] such as the Ca^{42} (p,p') and Ca^{44} (p,p') greatly aided this study in the information they provided on the levels of these two nuclei.

II. EXPERIMENTAL METHOD

The information on the levels of Ca^{42} and Ca^{44} was obtained by studying the decays of radioactive K^{42} (12.5 hr half-life) and Sc^{44} in the 0.26-Mev isomeric state (2.44 day half-life). The studies mainly consisted of measuring gamma-ray spectra in singles and coincidence, using NaI(Tl) scintillation spectrometers.

The K^{42} sources were reactor produced and obtained from Oak Ridge National Laboratories in the form of KCl dissolved in concentrated HCl. Preliminary experiments soon showed that a discernible Na^{24} contamination was present. It was in evidence by a very weak line at 2.76 Mev, having a half-life of Na^{24} (14.9 hr) rather than K^{42} (12.4 hr). The Na^{24} was present to about one part in 10^5. All subsequent sources routinely then had sodium chemically separated. The method of separation relied on the relative solubilities of sodium and potassium tetraphenylborate in 0.1 normal HCl. The sodium salt is extremely soluble, the potassium somewhat less so. Two or three filtrations of the potassium tetraphenylborate were carried out for each source, every filtration recovering about 75% of the potassium. There was no further evidence of any contamination in the precipitate and the line at 2.76 Mev was no longer detectable.

Sources of Sc^{44*} were obtained through the K^{41} (α,n) Sc^{44*} reaction by 40-Mev α-particle bombardments in the University of Washington cyclotron. Both the ground (3.9 hr half-life) and the isomeric states were produced, but the 3.9 hr activity was in equilibrium by the time the sources were used. The scandium was chemically separated by forming its thiocyanate salt and performing an ether extraction. Scandium thiocyanate, in contrast to the calcium and potassium salts, is much more soluble in ether than in acid, and could be extracted by the ether from the acid solution. For the gamma-ray measurements liquid sources were used that contained some sulfur residue (due to breakdown of the thiocyanate). For the beta-ray measurements the sulfur residue was removed completely by fuming with concentrated HNO_3 and $HClO_3$, until only scandium oxide remained. Additional $HClO_3$ then converted the oxide into the scandium perchlorate salt, which is soluble in water. The solution was transferred to the source backing (0.5 mil Al) and dried. There was no evidence of any other activities in these scandium sources except Sc^{43} (3.9 hr), which was completely undetectable after one day.

The singles work was done using a 3 in.×3 in. cylindrical NaI(Tl) crystal mounted on a Dumont 6364 photomultiplier, in conjunction with a 200-channel pulse height analyzer. The detector was housed in a 2-in. thick lead shield with a conical collimation hole in the roof. The sources were placed in a Lucite cup at

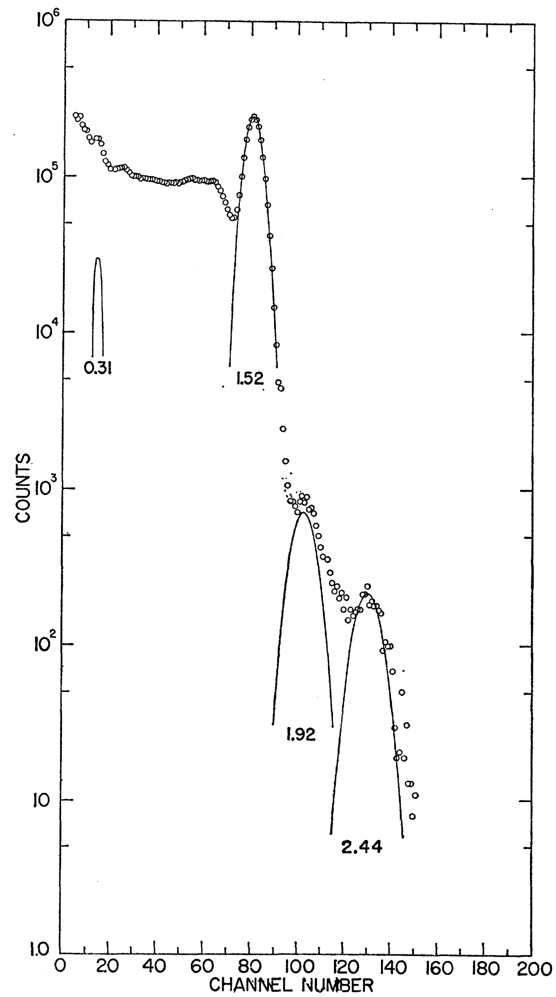

FIG. 1. Gamma-ray singles spectrum of the $K^{42} \rightarrow Ca^{42}$ decay.

[10] J. W. Blue and E. Bleuler, Phys. Rev. **100**, 1324 (1955).

[11] C. M. Braams, thesis, Utrecht, Holland, 1956 (unpublished); C. M. Braams, W. W. Buechner, and M. Mazari, Massachusetts Institute of Technology, Laboratory for Nuclear Science Progress Report, November 30, 1957. W. W. Buechner and M. Mazari, Rev. Mexicana Fis. **7**, 117 (1958).

the top of the hole 9.5 cm from the crystal. The source cups had a bottom thickness sufficient to stop any beta particles involved in the decays.

NaI crystals 2 in. diam×2 in. and $1\frac{3}{4}$ in. diam×2 in. were used with RCA 6810A photomultipliers for the coincidence measurements. A fast-slow coincidence arrangement gated the 200-channel pulse height analyzer. The fast coincidence circuit had 2τ equal about 35 mμsec. The counters were positioned at 90° about $1\frac{1}{4}$ in. from a Lucite source cup, with a 0.5-in. lead plate between the counters.

In addition to the gamma-ray work, a search was made for internal conversion electrons in the decay of Sc^{44*}. Preliminary measurements were taken with a solenoid-type lens spectrometer constructed[12] at the laboratory for certain electron polarization measurements. It had a resolution and transmission of about 5%. Later a more precise measurement was made with a double-focusing spectrometer which was designed and built by Bartlett. The design is similar to one constructed previously[13] and was used with a resolution of 1% and a transmission of 1%.

The scintillation counters were frequently calibrated during the course of the various singles and coincidences measurements. The sources used for this purpose were Cs^{137} (0.663 Mev), Co^{60} (1.17 and 1.33 Mev), Zn^{65} (1.12 and 0.511 Mev), Pr^{144} (2.18 Mev), and ThC″ (2.615 Mev). Several of these lines were also of aid in the decomposition of the spectra of K^{42} and Sc^{44}. The intensity measurements were made by measuring the peak height and the half-width of the Gaussian photopeak and then correcting for the intrinsic efficiency of the crystal and the photo to total ratio. Use was made for this work of empirically determined peak to total ratios for the particular crystals and geometries used here and the efficiencies calculated by others.[14–16]

III. EXPERIMENTAL RESULTS

A. Decay of K^{42}

The gamma-ray spectrum of K^{42} was measured a total of 15 times, each run representing from 10 to 100 min running time depending on the source strength. Background represented about a 25% correction to the photopeaks of the high-energy lines and was subtracted from each run. With one source the singles spectrum was followed through eight half-lives; no variation in the relative intensities of the lines was observed. Figure 1 shows a typical singles gamma-ray spectrum. Four lines are clearly identified, at 0.31±0.01 Mev, 1.52±0.01 Mev, 1.92±0.01 Mev, and 2.44±0.02 Mev. A decomposition of the K^{42} spectrum using single gamma-ray spectra from other sources yielded the photopeaks as shown. The intensities from this work are shown in Table I, and are in good agreement with the work of Morinaga, Mutsuro, and Sugawara.[7] The errors associated with the intensity measurements are considerably larger than the statistical errors of counting, due to the uncertainties in the peak to total ratios, the crystal efficiency, the subtraction processes and the fitting of a Gaussian distribution.

Weak cascades to the first excited state in general cannot be resolved in the singles spectra, because of the 1.52-Mev Compton distribution, and must be observed through coincidence measurements. A number of coincidence spectra were run with the window of the discriminator positioned on the photopeak of the 1.52-Mev gamma ray. Because of the very low intensity of

TABLE I. $K^{42} \rightarrow Ca^{42}$ gamma-ray intensities.

Gamma-ray energies (Mev)	Singles	Gamma-ray intensities (percent) Coincident with 1.52-Mev γ[a]	Coincident with 0.3-Mev γ[a]
0.31±0.01	1.1±0.1	1.1[b]	1.1[b]
0.49±0.02	···	<0.1	<0.1
0.60±0.02	···	0.06	0.2
0.90±0.02	···	0.1	0.1
1.02±0.02	···	0.1	0.2
1.52±0.01	100	···	···
1.92±0.01	0.4±0.1	0.3	0.3
2.44±0.02	0.2±0.1	···	···

[a] Errors on these intensities are discussed in the text.
[b] This value is taken from singles measurements, and serves as reference for the other coincidence intensities.

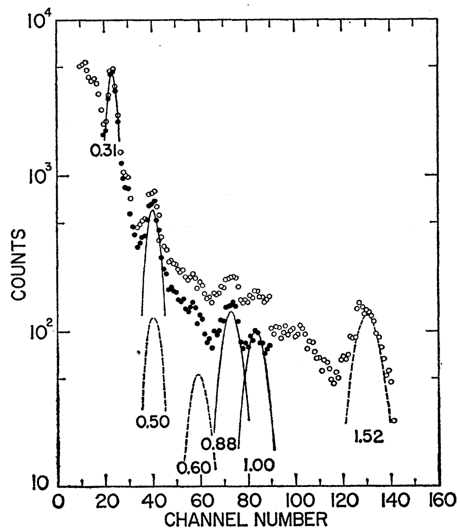

FIG. 2. Gamma-ray spectrum of $K^{42} \rightarrow Ca^{42}$ in coincidence with 0.4–1.6-Mev radiation. Open circles are the experimental points. Black circles are the result of a subtraction of chance coincidences. The dashed peaks at 0.50 and 0.60 Mev are taken from the 1.52-Mev coincidence spectrum.

[12] J. K. Kliwer and J. J. Kraushaar (unpublished).
[13] A. A. Bartlett and K. T. Bainbridge, Rev. Sci. Instr. 22, 517 (1951).
[14] G. White Grodstein, National Bureau of Standards Circular NBS-583.
[15] P. R. Bell, in *Beta- and Gamma-Ray Spectroscopy* (North-Holland Publishing Company, Amsterdam, 1955).
[16] R. L. Heath, Atomic Energy Commission Report TID-4500 (U. S. Government Printing Office, Washington, D. C., 1957).

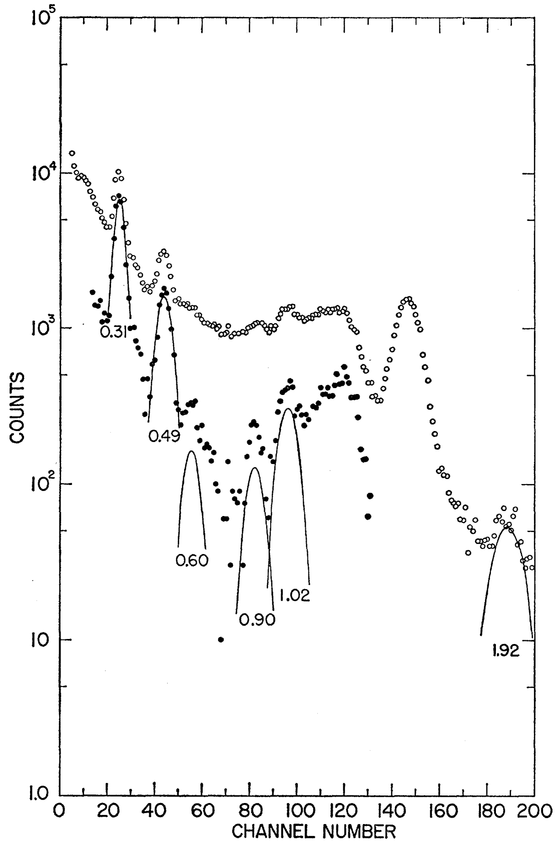

FIG. 3. Gamma-ray spectrum of $K^{42} \rightarrow Ca^{42}$ in coincidence with 0.2–1.4-Mev radiation. Open circles are the experimental points. Black circles are the result of the subtraction of chance coincidences.

the transitions of interest the number of chance coincidences in the spectra were sizable. Thus, in order to get a sufficient number of counts in the coincidence spectrum after subtraction of chances, long runs were required (1000 to 4000 min) and gain drifts were experienced. This difficulty made an intensity analysis of the 1.52-Mev coincidence runs marginal. Lines, however, were consistently seen at (0.31±0.01), (0.49±0.02), (0.60±0.02), (0.90±0.02), (1.02±0.02), and (1.92±0.02) Mev. In order to increase the counting rates, the window of the discriminator was opened up to cover the region of 0.4 to 1.6 Mev. Since the intensities of other gamma rays are so very low compared to the 1.52-Mev gamma ray, this widening of the acceptance window has a small effect on the coincidence spectrum. In Fig. 2 is shown a typical coincidence spectrum taken under these conditions. It was shown by calculation that the entire peak at 1.52 Mev was due to chances. A singles spectrum was thus matched to this peak and subtracted from the coincidence spectrum. The resulting spectrum was decomposed, yielding the photopeaks shown in Fig. 2 at 0.31, 0.50, 0.88, 1.00, and 1.92 Mev. The averaged energies and intensities derived from all data are shown in Table I. Since it was certain that the 0.31-Mev gamma ray was completely in coincidence with the 1.52-Mev gamma ray, the coincidence intensities have been normalized to the intensity of the 0.31-Mev line in singles (1.1%).

There is some question as to the reality of a gamma ray at 0.50 Mev. Annihilation quanta cannot clearly be resolved from the peak at 0.50 Mev, although the peak was consistently about 0.01 to 0.02 Mev lower than annihilation radiation at 0.51 Mev. It would be possible to have a 1.52-Mev gamma ray enter the discriminating counter and undergo pair production, leaving 1.0 Mev there and have a 0.51-Mev quantum escape to the other counter. It would also be possible for pair production to take place in the source or the nearby lead shield. It was felt, however, that the consistent appearance of lines at 0.49 and 1.02 Mev when the discriminator was positioned to accept just the full energy of the 1.52-Mev gamma ray was good evidence for the reality of these gamma rays. In such measurements, spurious annihilation events due to the 1.52-Mev radiation will not be seen, and only the high-energy gamma rays (1.92 Mev and 2.44 Mev) can lead to a spurious peak at 0.50 Mev. Such events are so highly improbable that they cannot reasonably account for the entire 0.50-Mev peak seen in the 1.52-Mev coincidences.

In Fig. 2 the 0.50-Mev line is greatly enhanced by annihilation radiation. Part of this enhancement is undoubtedly due to the spurious effects mentioned above. Part also may be due to the detection of internal pairs from the $0^+ \rightarrow 0^+$ decay of the 1.84-Mev state. The quantitative estimate of the contributions from the two effects is difficult, and only an upper limit (gained from the 1.52-Mev coincidence measurements) can be put on the 0.50-Mev gamma ray. This upper limit is shown as a dashed peak in Fig. 2. Also dashed is the peak at 0.60 Mev, which is not clearly resolved in this curve, but which was consistently seen with the indicated intensity in the 1.52-Mev coincidences.

A series of 0.31-Mev coincidence measurements were also undertaken to verify the positions of the various gamma rays in the decay scheme. Two types of measurements were taken; three runs were taken for gamma rays in coincidence with only the 0.31-Mev peak (the discriminator was set from 0.2 to 0.4 Mev) and for one run the discriminator was opened up to cover the region from 0.2 to 1.4 Mev. In all four of these runs the discriminator also was set on the Compton distribution of the 1.52-Mev gamma ray so that a mixed spectrum resulted. Those gamma rays, however, that are in coincidence with the 0.31-Mev gamma ray should be somewhat enhanced relative to the 1.52-Mev coincidence spectra. In the first three runs gamma rays were consistently seen at 0.31, 0.49, 0.60, 0.90, 1.03, 1.52, and 1.92. Mev. All of these lines were also seen in the 1.5-Mev coincidence spectra. Because of the low counting rates it was difficult with these measurements to determine intensities with any accuracy. To increase the counting rates the discriminator window was opened up to cover the region from 0.2 to 1.4 Mev. The results of these measurements are shown in Fig. 3, and the resulting intensities are shown in Table I. The broad peak just below 1.5 Mev in Fig. 3 was shown to be due to effects involving the back scattering of the 1.52-Mev gamma ray. The peak at 0.49 Mev, as with the 1.5-Mev coincidence measurements, contains a large contribution from annihilation radiation.

The errors on the intensities of the gamma rays observed in the coincidence measurements (which are shown in Table I) are large, mainly because of the weakness of the transitions and the uncertainties in the subtraction processes. There are also uncertainties in the intensity correction factors as well as statistical errors. The errors are difficult to meaningfully evaluate because of the many contributing factors, but the values listed can conservatively be taken to be valid within a factor of 2. That is, an intensity of 0.2% can be taken as having outside limits of 0.1 to 0.4%.

As a further check on the cascade position of the gamma rays, a run was taken in coincidence with all energies higher than 1.9 Mev, i.e., with the 2.44-Mev gamma ray. Counting rates for these runs were, of course, extremely low, and 4000 minutes were required for collection of the data shown in Fig. 4. The chances in this run were not large, as can be seen by the absence of a distinct peak at 1.5 Mev. Peaks at 0.48 and 1.00 Mev are apparent. It was not felt that the data represented in Fig. 4 were good enough to analyze for intensities, but it did serve to show that the 0.49- and 1.02-Mev lines were in coincidence with the 2.44-Mev gamma ray. Details of the decay scheme will be discussed later.

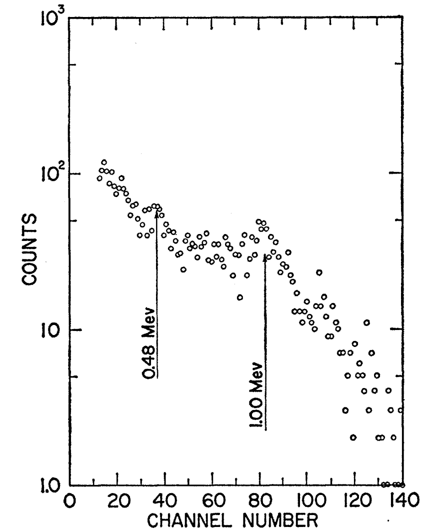

FIG. 4. Gamma-ray spectrum of $K^{42} \rightarrow Ca^{42}$ in coincidence with radiation >1.9 Mev.

B. Decay of Sc^{44}

A total of 10 measurements were made of the singles spectrum in the Sc^{44} decay, a typical one being displayed in Fig. 5. Observable in these spectra were the (0.26±0.01) Mev transition from Sc^{44*}, annihilation radiation, and a prominent gamma ray at 1.16 Mev. Weaker transitions above the 1.16-Mev line were resolvable at (1.50±0.02) Mev, (1.72±0.02) Mev, (2.28±0.02) Mev, and (2.69±0.02) Mev.

TABLE II. $Sc^{44}-Ca^{44}$ gamma-ray intensities.

Gamma-ray energy (Mev)	Gamma-ray intensity (percent) Singles (exp)	Singles (corrected for 1.12-Mev γ)	Coinc. with 1.16-Mev γ (rel. to 1.16-Mev γ singles)	Coinc. with 1.16-Mev γ (corrected for multiple triggering)
0.511±0.01	178	187	187[c]	187
0.68 ±0.02	...	...	3.2	3.2
1.02 ±0.02	...	...	6.2	3.1
1.12 ±0.02	...	...	9.4	4.7
1.16 ±0.01	100[a]	100[b]	...	...
1.50 ±0.02	1.7	1.8	1.7	1.7
1.72 ±0.02	0.8	0.8	0.7	0.7
2.28 ±0.02	0.2	0.2	...	...
2.69 ±0.02	0.2	0.2	...	...

[a] Includes 1.16-Mev and 1.12-Mev peaks.
[b] Includes only 1.16-Mev contribution.
[c] This value is taken from (corrected) singles measurements, and serves as reference for the other coincidence intensities.

Most of the apparent 1.72-Mev peak in Fig. 5 is due to the coincidence sum peak of the 1.16-Mev and 0.51-Mev radiations at 1.67 Mev. A comparison of the spectrum with that of Na^{22}, however, shows that the entire peak cannot be attributed to this effect. (From the coincidence summing point of view, Sc^{44} and Na^{22} are virtually identical sources; in both cases close to 90% of the feeding of the excited level is due to positrons, and the energies of the subsequent gamma rays are closely related.) After taking into account the slight difference in crystal efficiency for a 1.16-Mev gamma ray (Sc^{44}) and a 1.28-Mev gamma ray (Na^{22}), it was determined that (60±10)% of the apparent 1.72-Mev intensity in Fig. 5 was ascribable to coincidence summing. The entire summing contribution was assumed to affect the 1.72-Mev transition, leaving the 1.50-Mev line essentially unperturbed. This assumption was borne out by the result of coincidence measurements.

Intensities of the peaks seen in singles were measured in the same manner as for K^{42}, and are listed in Table II (with the 1.72-Mev line intensity corrected for summing contributions.)

Spectra were also taken in coincidence with the 1.16-Mev and 0.511-Mev gamma rays. The 1.16-Mev coincidence measurements were taken with the gate counter set from 1.0–1.4 Mev, and represent a total running time of 4000 min. In Fig. 6 the prompt coincidence spectrum is plotted with open circles, and the measured chance spectrum is shown as a dashed line. For purposes of analyzing the prompt coincidence spectrum, the chances were regarded as negligible.

The peak at (1.14±0.02) Mev in the spectrum is then

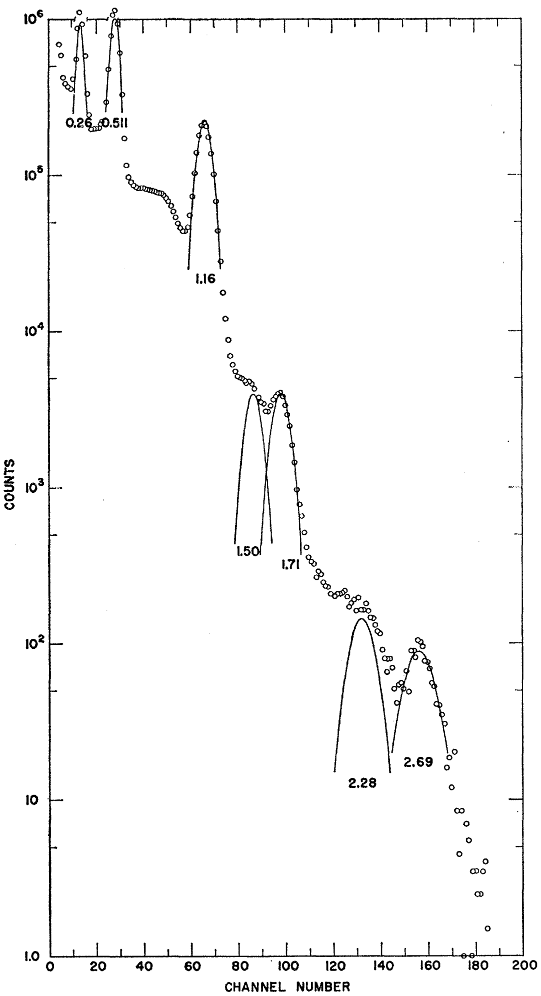

FIG. 5. Gamma-ray singles spectrum of the $Sc^{44} \rightarrow Ca^{44}$ decay.

only attributable to a new transition. Because a gamma ray of this energy will also trigger the gate counter, the apparent 1.14-Mev peak is actually a superposition of two peaks, the prominent 1.16-Mev transition, and a 1.12-Mev line in coincidence with it. Also in evidence in Fig. 6 are the 1.50-Mev and the 1.72-Mev lines; since the 1.16-Mev line is used to trigger the analyzer, sum effects are not measured, and the 1.72-Mev line is seen in its true intensity relative to the 1.52-Mev gamma ray. The only other resolvable lines are seen at (0.68±0.02) and 1.02±0.02 Mev, although unresolved lines between 0.70 Mev and 1.0 Mev may be present.

To obtain the intensities of these lines relative to the 1.16-Mev gamma-ray intensity, it was assumed that all positrons were annihilated in the source holder. The intensities of the coincidence lines were then measured relative to the annihilation radiation peak resulting from the positron decay. Since virtually all positron decays from Sc^{44} feed the 1.16-Mev first excited state, the intensities relative to the 0.511-Mev line in coincidence can be referred to the 1.16-Mev gamma-ray intensity by setting the 0.511-Mev intensity at 178%, its observed value in singles. This method of obtaining intensities, however, neglects the contribution of the 1.12-Mev gamma ray in singles. Its contribution may be estimated by the above procedure and subtracted from the 1.16-Mev singles intensity to provide a more accurate estimate of the other intensities. In Table II the coincidence intensities are listed with respect to the 1.16-Mev intensity (after correction for the 1.12-Mev contribution).

Additional corrections to the intensities must be made because of the fact that the 1.02-Mev, the 1.12-Mev, and the 1.16-Mev gamma rays can all trigger the gate counter. The 1.02-Mev gamma ray will be least effective in this regard, since only half of its photopeak is included in the gate setting. The gate can also be triggered by

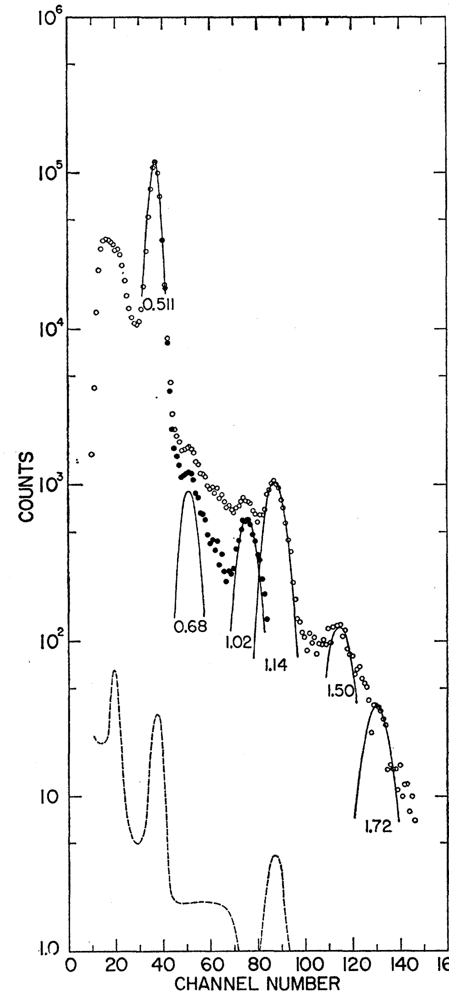

FIG. 6. Gamma-ray spectrum of $Sc^{44} \rightarrow Ca^{44}$ in coincidence with 1.0–1.4-Mev radiation. Open circles are the experimental points. Black circles are the result of a subtraction of the 1.14-Mev contribution fitted to the 1.14-Mev peak.

1.50-Mev and 1.72-Mev events which are partially absorbed by the crystal, but the contribution from such events will be rather small. The corrections necessitated by the multiple triggering depend on the positions of the gamma rays in the level scheme, and a discussion of their nature will be deferred until such assignments have been made.

The 1.72-Mev gamma ray was shown in Fig. 6 to be in coincidence with the 1.16-Mev radiation. Because of the low intensity of the line, this fact was not immediately apparent, and the possibility existed that it represented a crossover transition to the Ca^{44} ground state. Since such a level should then be fed by positrons, the possibility could most easily be eliminated by studying 0.511-Mev coincidences. Because the Compton distribution from the 1.16-Mev spectrum intrudes below the 0.511-Mev peak, the spectrum of Fig. 6 was superimposed on the 0.511-Mev coincidence spectrum. In Fig. 7, therefore, the spectrum of Fig. 6 has been fit, matching the respective heights of the 0.511-Mev lines. The subtracted curve, shown by the solid line, shows only a small 1.50-Mev peak, and no appreciable 1.72-Mev peak. Thus virtually the only state fed by the 0.511 Mev is the 1.16-Mev level, verifying the assumption made in the analysis of Fig. 6, and demonstrating that the 1.72-Mev line originates from a high-lying state.

Because of the occurrence of a 0^+ state as the second excited 1.84-Mev level in Ca^{42}, and because it was not clear whether an analogous state recurs in Ca^{44}, a search of the low-energy region for evidence of such a state was undertaken. If such a state occurs above the 1.16-Mev 2^+ level, and is populated, its major depopulation will be by a cascade transition. The state would not be populated by the positron decay (this would require a second forbidden transition) and could only be fed from higher populated states, such as the 2.28-Mev or 2.66-Mev states. No gamma rays consistent with such a decay were detected, making the existence of such a level improbable, although weak population of it cannot be excluded. If, however, the proposed 0^+ state were lower in energy than 1.16 Mev, a very small population would be detectable by observation of the conversion electrons; with no intervening states, and with a decay energy <1 Mev, the $0^+ \rightarrow 0^+$ transitions will proceed virtually completely by the internal conversion process.

In an effort to determine the existence of such a state, a search for conversion electrons was made with the double-focusing beta-ray spectrometer mentioned above. The high resolution, of which the machine is capable, was not employed for the search, since the sources were prohibitively weak, but the 1% resolution used was adequate to search for the lines. The conversion line at 0.22 Mev corresponding to the decay of Sc^{44*} to the Sc^{44} ground state was seen, and served as a convenient intensity calibration. No positive evidence for any other conversion lines up to 1.0 Mev could be seen; by using the observed intensity of the 0.22-Mev peak

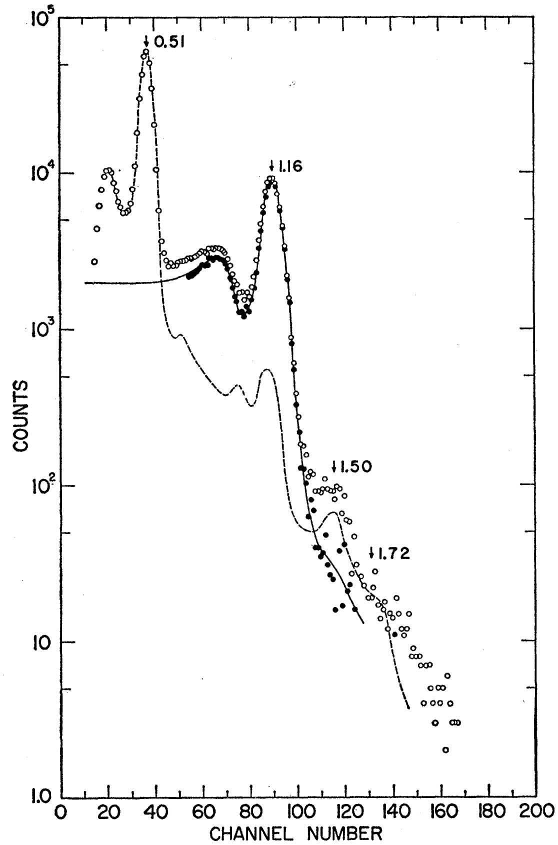

FIG. 7. Gamma-ray spectrum of $Sc^{44} \rightarrow Ca^{44}$ in coincidence with 0.511 Mev radiation. Open circles are the experimental points. Dashed line is a 1.16-Mev coincidence spectrum fitted to the annihilation peak. Solid line is the result of subtracting this contribution, and is the spectrum in coincidence with annihilation quanta only.

and the known conversion coefficient for the Sc^{44*} decay, an upper limit of 0.2% could be put on the percentage population of a state decaying wholly by internal conversion.

An independent upper limit was established by measuring the delayed coincidences between the possible gamma rays populating the state and the subsequent conversion electrons. For these measurements, the fast coincidence circuit was bypassed, and signals from the counter were fed directly to the slow circuit. To count electrons, the smaller NaI(Tl) crystal was removed and replaced by a Pilot-B plastic scintillator. All electrons with energies greater than 200 kev could trigger the analyzer gate. The input to the 200-channel analyzer was delayed 3 μsec relative to the electron pulses; since the slow circuit resolving time had $2\tau = 0.9$ μsec, prompt coincidence events were eliminated completely. To measure chance counting rates, the delay was placed in the electron counter circuitry, and the measurements repeated. The spectra so gained were virtually identical.

The upper limit on the feeding of the hypothetical 0^+ state was then gained by measuring the departure of the electron-delayed gamma ray coincidence spectrum from the chance spectrum. By such comparisons it was established that no gamma ray with energy between 0.2 and 2.0 Mev could feed such a state to greater than 0.05% of the total decay. The upper limit is contingent on the assumption that the lifetime of the state is between 10^{-5} and 10^{-7} sec. These are reasonable limits for the state's lifetime.

IV. LEVEL SCHEMES

A. Ca^{42}

To the right in Fig. 8 are shown the energies of the levels obtained from reaction studies.[11] The gamma rays reported here can all be accounted for as representing transitions between these levels. Clearly the 1.52- and 0.31-Mev lines are transitions from the first and second excited states. The spin sequence of 0-2-0 for these levels is consistent with the absence of a crossover transition. The 2.44-Mev gamma ray is not in coincidence with 1.52-Mev line, and must represent a direct transition to the ground state from the 2.42-Mev level. The 0.90-Mev line is then the cascade gamma ray to the 1.52-Mev level, and the 0.60-Mev line is a cascade gamma ray to the 1.84-Mev level. The coincidence measurements showed the 0.60 enhanced relatively in the 0.31-Mev coincidence runs, which substantiates this assignment.

The position of the 1.92-Mev gamma ray is restricted to a cascade transition from the 3.44- to the 1.52-Mev level, because it is in coincidence with the 1.52-Mev gamma ray and there is not enough energy available in the decay for any other assignment. The 1.02-Mev gamma ray, which is in coincidence with both the 1.52- and 0.31-Mev lines, is then the transition from the 3.44- to the 2.42-Mev level. The only unassigned gamma ray is the line at 0.49 Mev. Its energy is compatible with a transition between the 3.25- and 2.76-Mev levels.

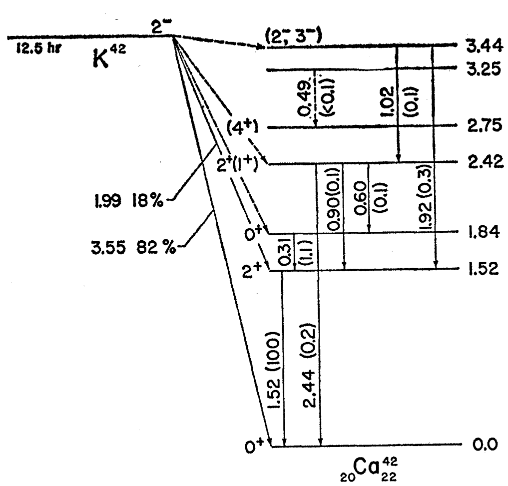

FIG. 8. Ca^{42} level scheme. Energies opposite the levels are those of Braams.[11] Intensities relative to the 1.52-Mev gamma ray are given in parentheses.

TABLE III. Log *ft* values of $K^{42} \rightarrow Ca^{42}$ decays. Branching ratios are computed by assuming 98.5% of the 1.52-Mev gamma ray is fed by 18% of the K^{42} decays. For transitions to the 1.84-Mev state the log f_1t values are given.

Energy levels (Mev)	Gamma rays feeding	Gamma rays leaving	Net feeding	Branching ratio	Log *ft*
1.52	0.31–1.1%	100%	98.5%	18%	7.5
	Higher–0.4%				
1.84	0.60–0.1%	0.31–1.1%	1.0%	0.18%	9.2
2.42	1.02–0.1%	2.42–0.2%	0.3%	0.05%	8.1
		0.90–0.1%			
		0.60–0.1%			
2.76	0.49– <0.1%				
3.25	None?	0.49– <0.1%	0.1%	<0.02%	>7.9
3.44	None	1.92–0.4%			
		1.02–0.1%	0.5%	0.09%	5.1

The lack of subsequent observed transitions from the 2.76-Mev level can partially be understood. The expected transitions would have energies of 0.34, 0.93, 1.23, and 2.76 Mev. The first two could be present to small percentages since they correspond with energies of other transitions. By assuming maximum contributions to the observed spectra, experimental upper limits of 0.1 and 0.05% have been placed on the presence of the second two transitions. From the spin assignments discussed later a 1.23-Mev transition is the most likely method of depopulating the 2.76-Mev level. Because of the uncertainty in the intensity of the 0.49-Mev gamma ray, the assignment as shown in Fig. 8 is not on a strong basis, but it is not incompatible with the data. The work of Morinaga *et al.*[7] is in general agreement with the level scheme in Fig. 8.

The spins and parities can be considerably restricted on the basis of the information shown in Fig. 8. The ground and first two levels have been discussed and have 0^+, 2^+, 0^+ for their spins and parities. The K^{42} ground state is a 2^- state; the spin 2 has been measured,[17] and the negative parity is required because of the first-forbidden character[6] of the ground state-to-ground state beta decay.

With the exception of the beta transition to the 3.44-Mev level, all others would appear to be first forbidden also. The spin of the levels then are restricted to positive parity and a spin $\leqslant 4$.

The 0.60- and 2.44-Mev transitions from the 2.44-Mev level to known 0^+ states require that the spin of that level be 1^+ or 2^+. The 1^+ assignment cannot be excluded experimentally, but systematics of other even-even nuclei in this region strongly point to a 2^+ state.

Very little can be said concerning the spin of the 3.25-Mev level. It is most probably populated almost entirely by a first forbidden beta branch, which would limit its spin and parity from 0^+ to 4^+. A 0^+, 3^+, or 4^+ might seem more favored because no gamma-ray transition to the ground state has been observed.

The ambiguous population of the 2.76-Mev level makes it difficult to assign a spin to it. The apparent absence of any transitions from it argue that it is fed

[17] E. H. Bellamy and K. F. Smith, Phil. Mag. 44, 33 (1953).

weakly, if at all, by beta decay. A spin of 4 is not unreasonable, but it could well be higher or possibly 0^+.

The log *ft* values of the various beta branches are also of aid in inferring spin assignments. In Table III are shown the calculated log *ft* values for the decays necessary to populate the states discussed above. The calculated log *ft* values are approximate since differences are in some cases being taken between intensities that have large errors. There is also the possibility that weak transitions which have not been observed could alter the branching ratio somewhat. The branching ratios should, however, in the most unfavorable cases be good within a factor of five and still permit the calculated log *ft* values to be used to discuss the forbidden nature of the beta transitions. The low log *ft* value for the 3.44-Mev state population indicates an allowed transition to it. This, coupled with the absence of any 3.44-Mev gamma ray, indicates a 2^- or 3^- assignment. All other log *ft* values are consistent with the assignments made; in particular the 0.1% upper limit on the 0.49-Mev gamma ray is consistent with a first forbidden decay to the 3.25-Mev level. One of the other close-lying states seen by Braams[11] between 3.25 and 3.89 Mev is almost certainly a 6^+ state. Such a state would not be populated in this decay, but systematics indicate its presence.

B. Ca^{44}

In the Sc^{44} decay, only one level has been previously identified, the first excited 2^+ state at 1.16 Mev. A second transition at 2.54 Mev has been reported,[10] which apparently corresponds to the 2.69-Mev line seen in this work. The good fit of the 2.69-Mev and 2.28-Mev lines with reaction data indicates that these are both crossover transitions and implies 2^+ assignments to both

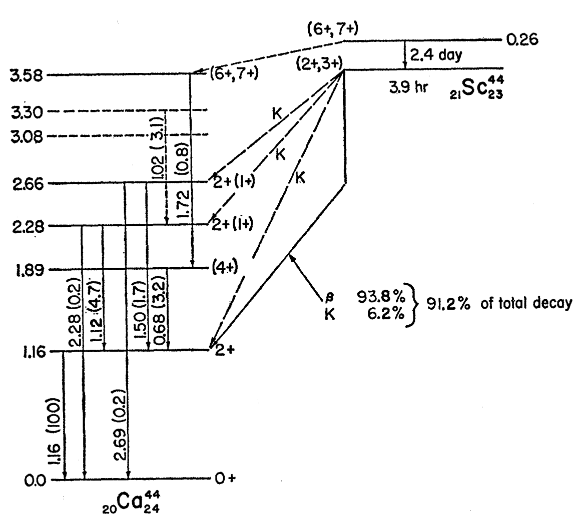

FIG. 9. Ca^{44}-level scheme. Energies opposite the levels are those of Braams.[11] Intensities relative to the 1.16-Mev gamma ray are given in parentheses.

TABLE IV. Log *ft* values in the $Sc^{44} \rightarrow Ca^{44}$ decay. Branching ratios from the Sc^{44} ground state are determined by assuming a total ground state population given by the 1.16-Mev intensity, plus the 2.28-Mev and 2.69-Mev crossovers, less the feeding of the 1.16-Mev level by the 1.72-Mev gamma-ray. "Net feeding" column is given with reference to the 1.16-Mev gamma-ray intensity (see Table II), while "branching ratio" column is normalized to total Sc^{44} ground state population. Log *ft* values are for the *K* capture branch. For the transition to the 3.61-Mev level, the 2.4-day half-life of Sc^{44*} was used.

Energy levels (Mev)	Gamma rays feeding	Gamma rays leaving	Net feeding by K and β^+	Branching ratio	Log *ft*
1.16	1.50–1.7% 1.12–4.7% 0.68–3.2%	1.16–100%	90.4%	90.7%	5.4
1.89	1.72–0.8% (others?)	0.68–3.2%	<2.4%	<2.4%	>5.6
2.28	1.02–3.1%	1.12–4.7% 2.28–0.2%	1.8%	1.8%	5.2
2.66		2.69–0.2% 1.50–1.7%	1.9%	1.9%	5.0
3.30		1.02–3.1%	3.1%	3.1%	3.8
3.61		1.72–0.8%	0.8%	0.8%	5.3

states. The 1.12-Mev and 1.50-Mev cascade transitions also fit well with these levels.

The 1.72-Mev line is energetically consistent with a transition from a 3.61-Mev level to the 1.89-Mev level seen by Braams.[11] Since there is evidence for a 3.58-Mev level in his work, it is most probable that this is the correct assignment for this transition. The 0.68-Mev transition can then be interpreted as the subsequent decay of the 1.89-Mev level to the 1.16-Mev state. In order, however, for the 3.61-Mev level to be so populated, the feeding must come from the metastable level in Sc^{44}, and hence requires a 6^+, 7^+ assignment for the state, the 6^+ being preferable from systematics. The 1.89-Mev level is then most logically a 4^+ state, which explains both the 1.72-Mev cascade and the lack of apparent population from the Sc^{44} ground state. It should be pointed out that this 3.6-Mev state is clearly not the same one seen by Cohen[18,18a] in his study of the decay of K^{44}. Braams[11] has seen several states of about this same energy, however, so that no contradiction exists. The 1.02-Mev transition is energetically consistent with a decay from the 3.30-Mev level seen by Braams, but this has difficulties, as will be described below. A decay scheme is shown in Fig. 9.

A check on the validity of these assignments can again be made by computing log *ft* values for the decays, but in order to do this, some of the measured intensities

[18] B. L. Cohen, Phys. Rev. **94**, 117 (1954).

[18a] *Note added in proof.* An experimental study of the decay of $K^{44} \rightarrow Ca^{44}$ has recently been reported (K. Sugiyama, T. Tohei, M. Sugawara, T. Dazai, and Y. Kanda, J. Phys. Soc. Japan **15**, 1909 (1960), which for the major part is consistent with our experiment. One discrepancy lies in their finding a state at 2.20 Mev (4^+) while we find one at 2.28 Mev (2^+). These may, however, be two different levels. A similar situation exists for their level at 2.55 Mev (2^+) which is in contrast to our level at 2.66 Mev (2^+). Our values of 2.28 and 2.66 Mev for these states agree well with the work of Braams.

must be corrected. The measured 1.14-Mev intensity is enhanced by the fact that either the 1.12-Mev or the 1.16-Mev gamma rays will trigger the gate counter, thus doubling the probability that a count will be recorded at this energy. Since the 1.02 Mev is in coincidence with both, its apparent intensity is also doubled, for the same reason. After these corrections are made, the log *ft* values can be estimated, and are listed in Table IV. The values are all consistent with allowed transitions, except for that to the 3.30-Mev level, which is abnormally low. This indicates either that the 1.02-Mev gamma ray's intensity is in error, or that it is incorrectly assigned in the decay scheme. The decay scheme assignment is not completely satisfactory, because of the lack of supporting evidence, but any alternative assignments require the introduction of levels not seen in (p,p') scattering, and also run into similar problems with low log *ft* values.

The ratio of positrons to 1.16-Mev gamma rays (1.16-Mev and 1.12-Mev gamma-ray composite) obtained was (see Table II) 0.89 which is in good agreement with a similar determination by Blue and Bleuler[10] (0.90±0.04), based on the relative intensities of the singles. They also obtained a value of $N_{\beta+}/N_{1.16}$ of 0.932±0.015 from a coincidence measurement between positrons and 1.16-Mev gamma rays. If one uses the theoretical K branching ratio of Zweifel[19] (6.2%) for the 1.47-Mev beta transition and the gamma-ray intensities reported here, one obtains a ratio of 0.86 for positrons to 1.16-Mev gamma rays.

V. DISCUSSION

The level schemes deduced from these experiments show quite clearly that a considerable modification of simple shell model predictions is necessary to account for the Ca^{42} and Ca^{44} spectra. The basic shell model predicts a $J=0, 2, 4, 6$ sequence for the lowest excited states of each nucleus, due to the coupling of the $f_{7/2}$ neutrons. It further predicts, because of seniority requirements, that the energy separation of the states are the same in both nuclei.

Recent theoretical work,[1–5] on the $f_{7/2}$ shell has tried to explain the failure of the shell model in terms of configuration mixing and of weak collective contributions. In the most recent shell model studies, Banerjee and DuttaRoy[2] have calculated the energy levels of Ca^{42} using the reaction matrix as the internucleon interaction, and Mitler[5] has made an empirical fit to the levels of Ca^{42} and Ca^{43} using a general interaction and configuration mixing. Both these studies indicate that the 2.44-Mev level in Ca^{42} is a 4^+ state, and neither account for the 1.84-Mev 0^+ state. The recent work of Raz[20] on the collective effects for a $(f_{7/2})^2$ configuration has partial success in fitting some of the levels seen in this experiment. He computed the variation of the energy levels as a function of the deformation parameter

[19] P. F. Zweifel, Phys. Rev. **96**, 1572 (1954).
[20] B. J. Raz, Phys. Rev. **114**, 1116 (1959).

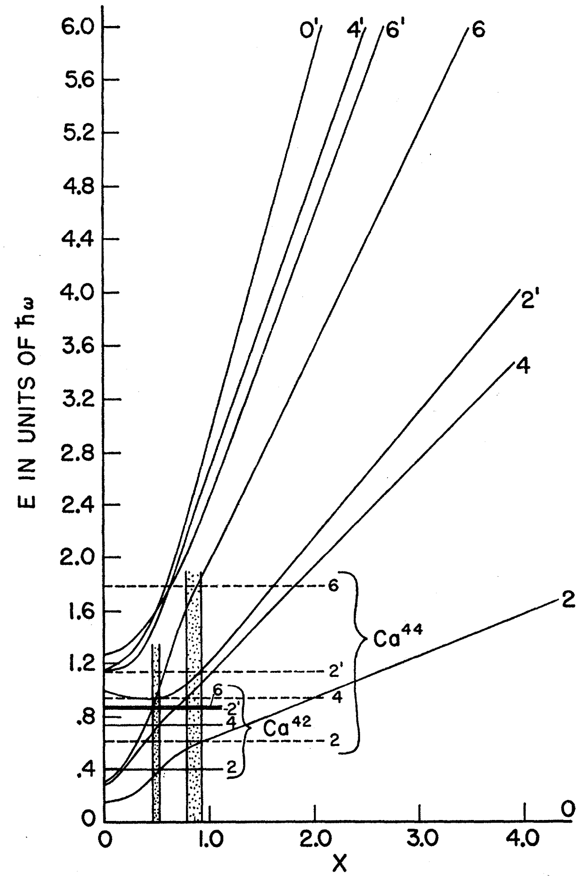

FIG. 10. Comparison of experimental levels with the collective model predictions of Raz.[20] Energy scale is in units of $\hbar\omega$. x is a parameter measuring the strength of the coupling. Solid lines are the states of Ca^{42}; dashed lines the states of Ca^{44}. Dotted areas indicate regions of best fit.

x and as a function of a parameter D that gives the relative strength of the two-body interaction. His curve for $D=0.2$ is reproduced in Fig. 10, with the pertinent energy levels of Ca^{42} and Ca^{44} superimposed. The choice of $D=0.2$ is not unique but appeared reasonable and calculations had been done at this value that could be compared with experiment. In these curves, the energies of the excited states are plotted as a function of the deformation parameter x. The energies are given in units of the vibration phonon energy $\hbar\omega$, and to fit the experimental data some value of $\hbar\omega$ must be determined. The fit for Ca^{44} was determined by matching the 6-0/2-0 energy ratio with the theoretical value. Thus the fit of the 4^+ 1.89-Mev and 2^+ 2.28-Mev states in Fig. 10 provide the test of the theory; the energy agreement for these states at $x=0.8$ is within 5%. For Ca^{42}, the fit was obtained by assuming that the 6^+ state and the second 2^+ state were virtually identical in energy. The energy of the first excited 2^+ state was then fixed at 1.52 Mev, and the 4^+ (2.76 Mev), $2'^+$ (assumed to be 3.25 Mev) and 6^+ (assumed to be 3.30 Mev) states were plotted on the appropriate scale. The 4^+ state fits quite well at $x \sim 0.5$, but the other two are overestimated by about 15% by the theory. The increase in x from Ca^{42} to Ca^{44}, and its modest value in either case, is what would be expected for these nuclei.

The energy values obtained are not, however, a critical test of the validity of the collective viewpoint. The fitting of the energy levels can also be accomplished by shell model calculations, if the strength of the interaction is left arbitrary, although the shell model cannot account for the second 2^+ state with any simple configuration. The most significant prediction of the collective theory, however, is that cascade transitions will be strongly enhanced relative to the competing crossover transitions. This effect is apparent in the experiments. In fact, the apparent enhancement is stronger than would be expected in virtually every case, although the uncertainty in intensities makes quantitative analysis marginal.

The apparent existence of collective effects does not suffice to explain all the observed states in Ca^{42} and Ca^{44}. In particular, the Ca^{42} 1.84-Mev and 2.42-Mev levels are disregarded. These could well be due to the elevation of two particles to the $p_{3/2}$ state, which from Ca^{41} data[21] are known to lie 1.9 Mev above the $f_{7/2}$ state. Such an excitation, however, would seem to require the $(p_{3/2})^2$ pairing energy to be abnormally high. The existence of a negative parity state as low as 3.44 Mev provides an argument supporting this effect. The recent measurement[22] of the lifetime of the 1.84-Mev level in Ca^{42} [mean life $=(4.8\pm0.3)\times10^{-10}$ sec] could well have a bearing on the correctness of the $(p_{3/2})^2$ configuration being important for this state.

If such $p_{3/2}$ configurations occur in Ca^{42}, one would naturally expect their recurrence in Ca^{44}. This may be the explanation of the 3.08-Mev level (unobserved in this work but seen in (p,p') measurements) and the 3.30-Mev level. A 0^+ state at 3.08 Mev would be difficult to detect, and cannot be excluded by this experiment. The 3.30-Mev level seems to be a 2^+ state.

If the 2^+ assignment is correct, the low $\log ft$ value of the decay feeding the 3.30-Mev state can be accounted for. The 1.16-Mev level is probably a recoupling of the $f7/2$ neutrons to $I=2$, with seniority 2; if the 3.30-Mev level is assumed to be the $I=2$, seniority 4 state, then the relative transition probabilities to these states from the Sc^{44} ground state can be calculated. P. S. Kelley and S. A. Moskowski [Z. Physik **158**, 304 (1960)] have performed such calculations; they find that $\log(|M_{GT}|^2{}_{\nu=4}/|M_{GT}|^2{}_{\nu=2})=1.4$. This is consistent with the observations in this paper; if one takes $\log ft=5.4$ for the decay to the 1.16-Mev state, the expected decay to a seniority 4 level would have $\log ft=4$.

[21] R. H. Nussbaum, Revs. Modern Phys. **28**, 423 (1956).

[22] P. C. Simms, N. Koller, and C. S. Wu, Bull. Am. Phys. Soc. **5**, 424 (1960).

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