Decay of $_{55}Cs^{129}$†

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The decay of $_{55}Cs^{129}$ [produced by the $(\alpha,2n)$ reaction on I^{127}] has been studied by use of the techniques of scintillation spectroscopy. The half-life of this activity was measured as 32.06±0.06 h. Gamma rays with energies 94, 174, 270, 278, 321, 375, 416, 549, 591, 904, and 945 keV have been observed in the singles and coincidence measurements. The 270-keV gamma ray was observed only in the coincidence measurements. Considerable effort was made to derive accurate energy and intensity information from the spectroscopic data. The electron-capture branching ratios to the ground state and excited states were determined from the coincidence measurements; log *ft* values for the various decay branches were derived from these ratios and the gamma-ray intensity measurements. A decay scheme is presented and compared with the pairing-model calculations of Kisslinger and Sorensen.

I. INTRODUCTION

THE decay of $_{55}Cs^{129}$ has been investigated in some detail by Jha *et al.*,[1] who employed techniques of scintillation spectroscopy ,and earlier by Nijgh,[2] who in addition measured the internal-conversion-electron spectrum by use of a beta-ray spectrometer. The low-lying levels in Xe^{129}, which are populated both by the decay of I^{129} and by the decay of Cs^{129}, have also been studied.[3–5]

In view of the recent pairing-model calculations of Kisslinger and Sorensen,[6] which included the level structures of the xenon isotopes, the authors felt that further clarification of the level scheme of Xe^{129} would be appropriate.

II. PREPARATION OF SOURCE

The 32-h Cs^{129} activity was produced by utilizing the $I^{127}(\alpha,2n)$ reaction. Natural iodine, in the form of calcium iodide, was irradiated by ~40-MeV alpha particles in the Argonne 60-in. cyclotron for a period of 4 h. The Cs^{129} obtained from this bombardment was sufficient to provide useful sources for several days. Following irradiation, the radioactive cesium was removed from the cyclotron target and chemically purified by means of a procedure developed by Hyde[7] for the isolation of francium. This procedure is based on the coprecipitation of cesium and silicotungstic acid in a solution saturated with gaseous hydrogen chloride. Cesium is separated from the redissolved precipitate by absorption on a cation exchange column. Elution of the column with concentrated HCl then yielded the carrier-free Cs^{129} activity.

Gamma-ray spectroscopic measurements performed on sources purified in this manner indicated the presence of no activities other than the Cs^{129}, with the exception of two scandium isotopes. A very weak gamma ray of energy 1157 keV is assumed to be the 1159±3-keV radiation from 3-day Sc^{44}. In addition, barely detectable amounts of 84-day Sc^{46} became observable in purified sources several weeks after preparation—by which time the cesium activity had decayed. Because of the extremely low intensity of these contaminants in the sources, it was felt that they would not interfere with the measurements, so further source purification was not attempted.

III. DETERMINATION OF HALF-LIFE

In order to establish that the observed gamma-ray spectrum did indeed represent the radiations of Cs^{129}, the spectral shape was recorded at intervals over several half-lives. A source was observed with a 2-×2-in. NaI(Tl) detector; care was taken that the experimental arrangement was not changed during the counting. Spectra taken at intervals during a 7-day period were carefully examined to reveal any change in spectral shape which would indicate the presence of contaminants with half-lives appreciably different from that of Cs^{129}. After subtraction of room background, no changes in spectral shape were observed during this series of measurements. The presence of Sc^{46}, noted previously, was detectable only after the cesium had entirely decayed.

The data described above were also analyzed to yield the half-life of Cs^{129}. For each of the spectra, the area between the valleys on either side of the 375–416-keV peak was determined. The values were least-squares fitted by computer to the usual exponential function. The value obtained for the Cs^{129} half-life is $T_{1/2}=32.06 \pm 0.06$ h. The quoted error was calculated by the computer on the basis of the deviations of the data points from the fitted function. The value is in satisfactory agreement with previous measurements with lower precision.[2,8]

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[1] S. Jha, H. G. Devare, and R. M. Singru, Nuovo Cimento **18**, 1108 (1960).

[2] G. J. Nijgh, thesis, University of Amsterdam, 1955 (unpublished).

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[6] L. S. Kisslinger and R. A. Sorensen, Rev. Mod. Phys. **35**, 853 (1963).

[7] E. K. Hyde, J. Am. Chem. Soc. **74**, 4181 (1952).

[8] R. W. Fink and D. H. Templeton, J. Am. Chem. Soc. **72**, 2818 (1950).

IV. MEASUREMENTS OF THE GAMMA-RAY SPECTRUM

For detailed analysis, the gamma radiations of Cs^{129} were observed with a collimated 4-×4-in. NaI(Tl) scintillator coupled to an EMI-9479A photomultiplier. The lead collimator, which is covered with a graded x-ray filter on all surfaces exposed to the detector, considerably increases the peak-to-total ratio obtainable with a given crystal. For the crystal described above, the height of the Compton distribution relative to that of the photopeak (at 662 keV) is approximately halved when the collimator is used. This improvement is very helpful in separating the low-energy transitions from the continuum of the intense 375-keV radiation from Cs^{129}. The resolution of the detector is about 7.2% at 662 keV.

The Cs^{129} gamma-ray spectrum obtained with a 512-channel pulse-height analyzer is shown in Fig. 1. Immediately before and after the Cs^{129} spectrum was accumulated, spectra of a number of accurately known γ-ray standards were recorded to provide energy and spectral shape calibrations for the spectrometer system. The isotopes used for this purpose were Na^{22}, Mn^{54}, Co^{60}, Cs^{137}, Re^{188}, and Hg^{203}. The photopeak of each of these calibration standards was fitted by a Gaussian function to determine its centroid and resolution. The fitting was accomplished by the use of a program, PHY 1724A written in FORTRAN for the IBM-704 computer. This program employs the variable metric method of minimization developed by Davidon.[9] A spectral background is established by constructing a cord between the minima immediately above and below the photopeak. This background is then subtracted and the Gaussian function least-squares fitted to the residual data points. All parameters involved in the fit, together with their calculated errors, are tabulated by the computer.

In order to establish the correspondence between the centroid X of the full-energy peak and the gamma-ray energy E, it was assumed that these variables were related by the quadratic function

$$E=A_0+A_1X+A_2X^2.$$

The coefficients are determined by fitting the function to the centroid values associated with the standard gamma rays listed above. Typically, $A_1/A_2\approx 10^4$. Above 100 keV, this simple relation has been found to fit the calibration points with deviations of less than 1 keV.

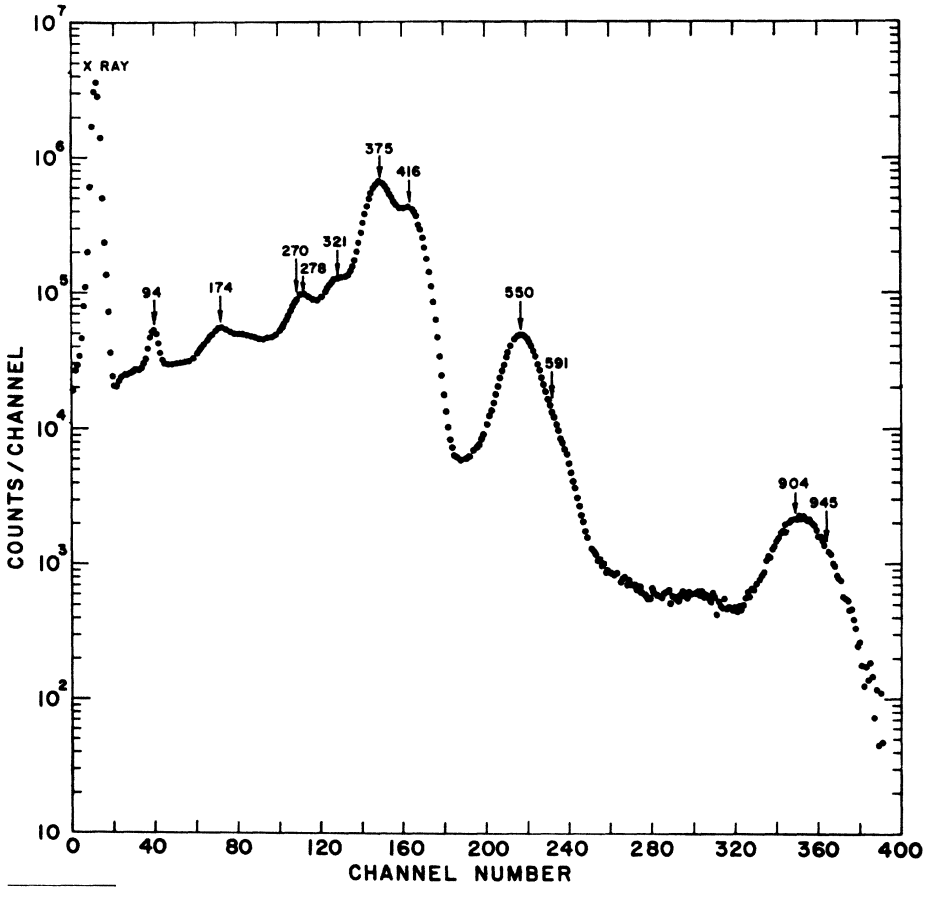

FIG. 1. Collimated gamma-ray singles spectrum detected with a 4-×4-in. NaI(Tl) crystal. Except in the region above channel 360, the experimental uncertainties are smaller than the size of the plotted data points.

[9] W. C. Davidon, Argonne National Laboratory Report No. ANL-5990, 1959 (unpublished).

The spectrum in Fig. 1 was analyzed by computer with the aid of spectral line shapes obtained from the standard sources mentioned above. The spectrometer response at some selected energy is synthesized by linearly shifting and expanding the nearest monochromatic standard shape to obtain the desired photopeak centroid with the appropriate half-width. The intensity of each synthesized shape is then adjusted by the computer so that the sum of the components is least-squares fitted to the data. The residual spectrum is then examined and the positions of the centroids are appropriately adjusted. The relative intensities of the gamma rays are then calculated from the computed amounts of the various constituent spectral shapes used in fitting the data. In the present measurements the difficulties of analysis were intensified by the fact that the majority of the gamma transitions were seen as incompletely resolved pairs. This feature of the data causes χ^2 (which is normally used to indicate the quality of the fit) to change only slowly as the centroids and the intensities are adjusted together. Considerable effort was made to derive the maximum information from these unresolved peaks.

In order to include the effects of collimation, the corrections for detection efficiency of the crystal were calculated by use of a Monte Carlo calculation developed by W. F. Miller and W. J. Snow.[10]

The results of the analysis of the gamma-ray spectrum, corrected for detector efficiency, are shown in Table I. The quoted error in the energy of each of the unresolved lines is based on the variation in the final centroid values obtained when the fit was started from a variety of different initial conditions. The errors calculated by the computer are usually smaller than the values assigned in Table I.

The region of the spectrum between 700 and 900 keV is of particular interest. The intensity of the spectral continuum here is larger than can be explained simply by the Compton distributions of the higher energy photopeaks. Jha *et al.*[1] interpreted this as indicating the presence of a 790-keV gamma transition, to which they assign an intensity of 0.15 relative to the 375-keV transition. Our measurements indicate that a single gamma ray in this region would have an intensity less than 0.05. Instead, we interpret the distribution in this region as resulting from the bremsstrahlung produced by electron capture to the lower states of Xe^{129}. Approximate analysis indicates a bremsstrahlung spectrum characteristic of a total decay energy of about 1150 keV. Such a spectrum was used as one of the components in the computer analysis of the gamma-ray distribution.

Due to the high internal conversion of the transition between the 40-keV first-excited state and the ground state, this gamma ray is not resolved from the intense xenon K x ray.

TABLE I. Gamma rays observed in the decay of Cs^{129}. Intensities are specified relative to a value of 100 for the 375-keV transition.

Energy (keV)	Intensity
94.5±1	1.32 ±0.1
174.5±2.5	1.02 ±0.1
270 ±3	0.78[a]±0.2
278.4±2	6.4 ±0.6
321.4±2	7.6 ±0.6
375.0±1.5	100
416.1±3	52 ±3
549.9±2	9.4 ±1.0
591.5±5	1.7 ±0.3
904.2±7	0.54 ±0.1
945.1±9	0.18 ±0.04

[a] Energy and intensity evaluated from γ-γ coincidence data.

[10] W. F. Miller and W. J. Snow, Argonne National Laboratory Report No. ANL-6318, 1961 (unpublished).

TABLE II. Experimentally observed γ-γ coincidences (indicated by x's). Except for the 904- and 945-keV transitions, all gamma rays were also seen in coincidence with the x ray. Row and column headings are energies in keV.

	416	375	321	278	270	174
94			x	x		x
174	x	x	x	x		
270			x	x		

V. GAMMA-GAMMA COINCIDENCE EXPERIMENT

The coincidence relationships between the various gamma rays resulting from the decay of Cs^{129} were investigated by use of two 2-×2-in. NaI(Tl) detectors. The crystals were arranged with an angle of 60° between their axes and with a $\frac{1}{2}$-in. lead antiscattering shield placed between them. Coincidences were detected by a fast-slow coincidence circuit with resolving times (2τ) of 50 nsec and 3 μsec, respectively.

The coincident pulses from the two detectors were recorded simultaneously by a 4096-channel two-parameter analyzer. Supplementary circuits have been constructed to permit the analyzer to store singles spectra from each of the detectors simultaneously with accumulation of the coincidence data. Either 100% or 10% (to reduce dead time losses) of the singles pulses from each of the counting channels is stored in the plane corresponding to the channel zero of each of the two analog-to-digital converters.

Most of the coincidence data were collected with the gains of the detection system adjusted so that the 64×64 storage matrix of the pulse-height analyzer covered the energy range 0–730 keV in each detector. Coincidence data were accumulated for approximately 36 h in this configuration. The source strength was maintained at an optimum level by occasional addition of source material.

The results of the coincidence measurements are displayed in Table II. Of particular interest is the identification of the 270-keV gamma transition which is not resolved from the more intense 278-keV radiation in the singles data. Evidence for the 270-keV transition

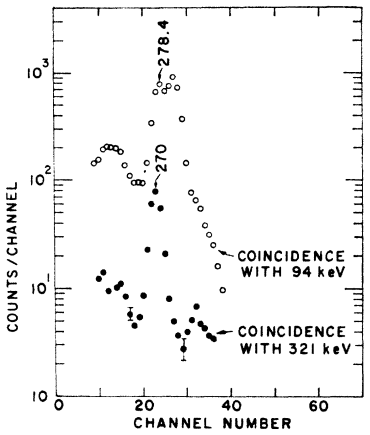

FIG. 2. Spectra taken in coincidence with the 94- and 321-keV gamma rays show evidence for the 270-keV transition. Errors on the upper curve are smaller than the data points.

was obtained by comparing slices through the coincidence matrix corresponding to spectra in coincidence with the 94- and 321-keV transitions (Fig. 2). Comparison of these spectra clearly shows the existence of two distinct transitions in the 275-keV region; the separation between the peak centroids of the two spectra was used to evaluate the energy of the 270-keV radiation.

Transitions are seen from each level (with the exception of the weakly populated state at 945 keV) to all other levels that are energetically available. Coincidences involving radiations with energies greater than 700 keV were sought but none were observed—presumably because of the low intensity of these transitions.

Previous measurements of the Cs^{129} decay[1] indicated the presence of several coincidence relations not observed in the present study. From the reported energies, at least some of these observed coincidences may have been due to scattering from one detector to the other rather than to coincident radiations from the source.

VI. MEASUREMENTS OF THE ELECTRON-CAPTURE BRANCHING

Knowledge of the electron-capture branching to the ground state and 40-keV first excited state of Xe^{129} is necessary to calculate log ft values for the various decay branches from Cs^{129}.

The number of K x rays per 375-keV photon was accurately measured. In addition, the ratio of x-x coincidences to x–375-keV-gamma coincidences was experimentally determined. From the results of these two measurements, values for the relative electron-capture branching to the ground state and the first excited state can be determined. To measure the intensity of the 29-keV K x ray of xenon, the photon spectrum of Cs^{129} was obtained with particular attention to the detector efficiency for low-energy radiation.

Because of the uncertainty in the x-ray attenuation by the window of the 3-×3-in. NaI(Tl) detector used for this measurement, the window transmission was determined empirically by use of a series of sources for which the relative intensities of x rays and gamma rays are known. These sources were mounted in the same way as the Cs^{129} under study, i.e., in a sandwich between two layers of 14-mg/cm² cellulose tape, and were counted on the crystal axis, 10 cm from the front face.

The window attenuation A_x for the particular K x-ray energy was then obtained by measuring the ratio of the areas N_x/N_γ, of the x-ray and gamma-ray photopeaks, i.e.,

$$A_x=\frac{N_x}{N_\gamma}\frac{1}{\alpha_K}\frac{R_\gamma}{R_x}\frac{E_\gamma}{E_x}\frac{A_\gamma}{\omega_K}, \tag{1}$$

where α_K is the K internal-conversion coefficient and ω_K is the fluorescence yield. Values for the ratio R of photopeak to total area and the detection efficiency ϵ were taken from the calculations of Miller and Snow.[10] Correction of photopeak area for escape of iodine x rays was based on the results of Axel.[11] On the basis of the measured intensity ratio of the 22.1-keV K x ray of Cd^{109} relative to the 89-keV gamma ray,[12] the thickness of the detector window was found to be equivalent to 183 mg/cm² of aluminum. This result is consistent with the actual detector window which consists of ~0.100 in. of MgO backed by a 0.019-in Al foil. The ratio of the number N_x of Xe^{129} K x rays to the number N_γ in the composite (375- and 416-keV) gamma peak was measured and corrected as indicated above. The efficiency used for this composite gamma peak was a weighted mean of the values appropriate to the two energies. The value obtained for the x-ray/γ-ray ratio is

$$N_x/N_\gamma=1.76\pm0.09, \tag{2}$$

where $N_\gamma=N_{(375+416)}$. With the use of this ratio, the electron-capture branching to the two lowest states in Xe^{129} can be evaluated. The average number N_x of x rays produced per decay can be related to the individual numbers N_i of electron-capture decays leading to level E_i by the expression

$$N_x=\left[\sum_i \omega_K N_i \sum_j \frac{f_{ij}}{\beta_i}\frac{\alpha_K(ij)}{1+\alpha(ij)}+\left(\frac{K}{T}\right)_i\right], \tag{3}$$

where f_{ij} represents the fraction of direct transitions from level i to level j, $\alpha_K(ij)$ and $\alpha(ij)$ are the K-shell and total internal-conversion coefficients for the transition $i\rightarrow j$ respectively, the factor $(K/T)_i$ represents the ratio of K-capture to total electron-capture for decays leading to level i, β_i is the ratio of direct beta-decay population to total population for level i, and the summations over i and j extend over all levels.

In Eq. (3), the factors N_i, f_{ij}, and β_i for all states above 70 keV were evaluated from the gamma-ray

[11] P. Axel, Brookhaven National Laboratory Report No. BNL-271, 1953 (unpublished).

[12] A. H. Wapstra and W. van der Eijk, Nucl. Phys. 4, 325 (1957).

intensity measurements. Values for the K-shell internal-conversion coefficients for the various transitions were taken from Rose[13]; all transitions were assumed to have the minimum multipolarity allowed by the spin assignments indicated in Fig. 3. Substitution of the measured value of N_x/N_γ into Eq. (3) provides a relation between the branches to the various levels of Xe^{129}. A similar relation that does not involve the electron-capture branch to the ground state can be obtained by again measuring the ratio of x rays to the combined 375- and 416-keV gamma rays and, in addition, by requiring that each radiation be in coincidence with the 29-keV K x ray.

This coincidence ratio was obtained from a separate measurement with the gamma-gamma coincidence apparatus described above. Repeating the experiment with several different coincidence resolving times demonstrated that the coincidence circuitry was fully efficient for the low-energy x-x coincidence. After correcting for the detector response (in the same manner as described for the 3×3-in. crystal) and for the asymmetry of the apparatus for the low-energy coincidence, the ratio of the number N_{xx} of x-x coincidences to the number $N_{x\gamma}$ of coincidences between the x ray and the combined 375- and 416-keV gamma peak was found to be

$$N_{xx}/N_{x\gamma}=0.42\pm0.04. \quad (4)$$

In the notation of Eq. (3), the number of x-x coincidences per decay can be written

$$N_{xx}=\omega_K^2\sum_i^{\text{levels}} N_i\left(\frac{K}{T}\right)_i \times\left\{f_{i,40}\left(\frac{\alpha_K(40)}{1+\alpha(40)}+\frac{\alpha_K(i,40)}{1+\alpha(i,40)}\right)\right\}. \quad (5)$$

Similarly, the number $N_{x\gamma}$ of coincidences between the x rays and the combined 375- and 416-keV gamma peak is

$$N_{x\gamma}=\omega_K N_{416}\left\{f_{416,40}\left[\left(\frac{K}{T}\right)_{416}+\frac{\alpha_K(40)}{1+\alpha(40)}\right]+f_{416,0}\left(\frac{K}{T}\right)_{416}\right\}. \quad (6)$$

By combining Eqs. (3)–(6) and substituting the experimental gamma-ray intensities, it is possible to place upper limits on the branching ratios for decays to the ground state and the 40-keV state. These ratios are

$$N_0/N_{416}=0.17_{-0.17}{}^{+0.28},$$
$$N_{40}/N_{416}=0.032_{-0.032}{}^{+0.084}.$$

[13] M. E. Rose, *Internal-Conversion Coefficients* (North-Holland Publishing Company, Amsterdam, 1958).

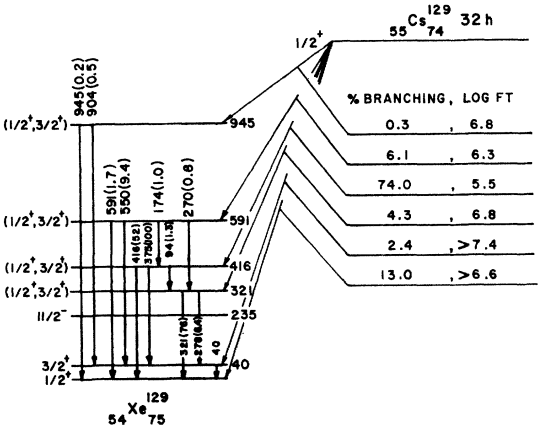

FIG. 3. Decay scheme for $_{55}Cs^{129}$ (32 h).

The values of log ft for the various beta-decay branches were derived from these ratios and are shown in Fig. 3.

VII. DISCUSSION

The present study confirms the general features of the decay scheme proposed by Jha *et al.*[1] The energies of the various levels presented in Fig. 3 have been adjusted to best fit the gamma-ray energy measurements listed in Table I. The gamma-ray singles and coincidence studies show the existence of, and appropriate coincidences among, transitions between each level and all other levels that are energetically available, the only exception being the weakly populated level at 945 keV. No evidence was found for the existence of a 720-keV state reported by Jha *et al.*[1]

The ground-state spins of I^{129}, Xe^{129}, and Cs^{129} have been measured by direct methods; the values are $\frac{7}{2}+$, $\frac{1}{2}+$, and $\frac{1}{2}+$, respectively.[14] The assignment $I=\frac{3}{2}+$ for the 40-keV level is based on the measured lifetime of this level and on the second-forbidden beta transition to it from I^{129}. The log ft values of all transitions from Cs^{129} to the higher states in Xe^{129} indicate allowed or first-forbidden character. The K internal-conversion coefficients of the gamma transitions in Xe^{129} as determined by Jha *et al.*[1] indicate that none of these transitions have multipolarity $E1$. This indicates that if the levels at 321, 416, and 591 keV have negative parity, then they must have a spin of $\frac{7}{2}$ or higher. However, such a high spin is not consistent with the log ft values observed for the electron-capture decay to these levels. We therefore conclude that these levels have positive parity. The electron-capture decays from the ground state of Cs^{129} must therefore be allowed and the spins of the higher states in Xe^{129} are restricted to $\frac{1}{2}+$ or $\frac{3}{2}+$.

It is interesting to note that the log ft values for beta decay to the various states in Xe^{129} seem to be consistently larger than the values usually associated with

[14] *Nuclear Data Sheets*, complied by K. Way *et al.* (Printing and Publishing Office, National Academy of Sciences–National Research Council, Washington 25, D.C.), NRC 61-1-96.

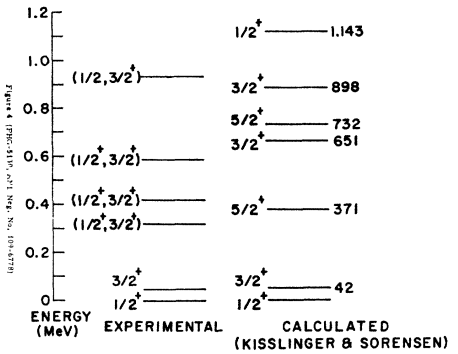

FIG. 4. Comparison of experimental data with results calculated by Kisslinger and Sorensen.

allowed beta decay. The authors see no explanation for this behavior except that the initial state appears to have a large admixture of the $\frac{5}{2}$ quasiparticle configuration.[6]

The recent pairing-model calculations of Kisslinger and Sorensen indicate (Fig. 4) that the six states with spin $\frac{1}{2}$ or $\frac{3}{2}$, obtained by coupling the available quasiparticles to a single phonon, lie below 1.4 MeV. A seventh low-spin state that was indicated by previous results[1] would suggest the need for introduction of additional quasiparticle configurations in calculating the low-energy spectrum; but the existence of this is not confirmed in the present study. Kisslinger and Sorensen predict a $\frac{5}{2}$ state at about 350 keV. Such a level would not be populated directly from Cs^{129} but one might expect to see gamma radiation to it from the higher xenon states of lower spin. No such gamma rays have been observed.

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Level Structure of I^{131}

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The levels of I^{131} excited in the decay of the 30-h activity of Te^{131m} have been studied. Scintillation spectrometers were used for recording the gamma spectrum and gamma-gamma coincidences. The internal-conversion spectrum with a double-focusing spectrometer showed K and L lines corresponding to gamma rays of energies 81, 102, and 150 keV and K lines corresponding to 200-, 241-, 336-, 452-, 775-, and 854-keV gamma rays. K conversion lines of 775-, 786-, 797-, 831-, 854-, 869-, 1127-, and 1206-keV gamma rays were observed in the external-conversion spectrum. The relative intensity of the gamma rays was found by analyzing the gamma spectrum and also from the external-conversion spectrum. By use of these relative intensities and the intensities of the internal-conversion lines, the conversion coefficients and the possible multipolarities of the gamma transitions were ascertained. In particular, the 200- and 241-keV transitions were found to be of $E1$ type, indicating the presence of odd-parity states in I^{131}. These two transitions were also found to be highly retarded and the 1829-keV level from which they arise was found to have a half-life of 5.9 ± 0.2 mμsec from delayed-coincidence measurements using a time-to-amplitude conversion technique. From these measurements it is concluded that Te^{131m} decays by beta transitions mainly to the 2012-, 1981-, 1965-, 1931-, and 1902-keV levels of I^{131}. These levels de-excite by transitions to the levels at 1829, 1629, 1583, 1340, 1145, 1065, 797, 775, 602, and 150 keV. From the beta spectrum studied with an intermediate-image spectrometer, it was concluded that the beta transition to the ground state has an end-point energy of 2460 ± 15 keV and a relative intensity of 6%, while the isomeric transition takes place with a relative intensity of 18%.

I. INTRODUCTION

THE levels of I^{131} excited in the decay of the 30-h activity of Te^{131m} have been studied by Hebb[1] and by Badescu *et al.*[2,3] It is known that Te^{131m} has a decay energy of over 2.4 MeV and feeds levels of I^{131} up to about 2 MeV. It also decays by the 182-keV isomeric transition, and the Te^{131} so formed subsequently excites by beta decay the levels of I^{131}. The decay scheme of this 25-min activity of Te^{131} has already been described earlier.[4] In this paper, we report the results of a detailed investigation of the properties of the levels

[1] Elizabeth Hebb, Phys. Rev. **97**, 987 (1955).
[2] A. Badescu, K. P. Mitrofanov, A. A. Sorokin, and V. S. Shpinel, Izv. Akad. Nauk SSSR, Ser. Fiz. **23**, 1434 (1959).
[3] A. Badescu, O. M. Kalinkina, K. P. Mitrofanov, A. A. Sorokin, N. V. Farafontov, and V. S. Shpinel, Zh. Eksperim. i Teor. Fiz. **40**, 91 (1961) [English transl.: Soviet Phys.—JETP **13**, 65 (1961)].
[4] S. H. Devare, P. N. Tandon, and H. G. Devare, Phys. Rev. **131**, 1750 (1963).