neutron scattering resulting in the excitation of purely vibrational states in deformed nuclei would be of value.

In summary, neutron elastic- and inelastic-scattering cross sections have been measured for W^{182}, W^{184}, W^{186} in the incident energy range 0.3 to 1.5 MeV. The results have been successfully analyzed by means of an interaction model containing a spheroidal optical potential, modified Hauser-Feshbach theory, and strong coupling between the ground state and the first excited state.

ACKNOWLEDGMENTS

The authors are indebted to a number of individuals at Atomics International and the Argonne National Laboratory for assisting various facets of this work. They are also particularly indebted to the National Aeronautics and Space Administration for making available the isotopically separated tungsten samples without which this work would have been impossible.

PHYSICAL REVIEW VOLUME 162, NUMBER 4 20 OCTOBER 1967

Gamma-Ray Spectrum of Ruthenium-99†

G. A. Moss* and D. K. McDaniels
Physics Department, University of Oregon, Eugene, Oregon
(Received 9 February 1967)

The γ-ray spectrum resulting from the decay of 16.1-day Rh^{99} to the excited states of Ru^{99} has been investigated using Ge(Li) solid-state detectors. The source, produced by the $Ru^{99}(p,n)Rh^{99}$ reaction using enriched Ru^{99} metal, was found to emit a large number of γ rays. The energies of the stronger γ rays (with their relative intensities) are: 89.4 (100), 175.2 (7.6), 322.4 (21.7), 353.0 (107.1), 442.8 (5.7), annihilation radiation (30), 527.7 (132.0), 618.0 (13.1), 941.5 (5.0), 1292.1 (1.2), 1532.9 (1.8), 1572.4 (7.7), and 1970.3 (0.5) keV. A number of lines not previously reported have been found. Coincidence measurements and γ-γ directional correlations have been performed using a 6-cc Ge(Li) detector in conjunction with a gain-stabilized NaI(Tl) spectrometer. The decay scheme has been constructed with the aid of coincidence studies and high-precision γ-ray energy measurements. The spin of the 618.0-keV level has been shown to be $\frac{1}{2}$, whereas the results of the 89.4–353.0 and the 175.2–353.0-keV correlations indicate either a $\frac{1}{2}$ or $\frac{3}{2}$ assignment to the 442.8-keV level. Attempts to find a theoretical model which represents the experimental data have been made, incorporating the Nilsson model, the intermediate-coupling model of Bohr and Mottelson, and the core-excitation model of de-Shalit.

I. INTRODUCTION

RHODIUM 99 decays by positron emission and electron capture to the excited states of ruthenium 99. The resulting γ-ray spectrum is very complicated and has thus hampered a complete study of the decay scheme. There has been very little experimental data published about the γ-ray spectrum of Ru^{99}. The available information has been restricted to two conference reports: Kurbatov, Townley, Feigley, and Kurbatov,[1] who made use of scintillation detectors to investigate the γ-ray spectrum and a magnetic spectrometer to study internal-conversion electrons of both the 16.1-day and 4.3-h isomers of Rh^{99}; and Jha, Johnston, and Nainan,[2] who reported on a study of the decay of Rh^{99} in which they had formulated a decay scheme utilizing scintillation spectroscopy. This information together with a "private communication" has led to the decay scheme presently found in the Nuclear Data Sheets.[3] More recently Connors[4] has made use of semiconductor detectors to study the states of Ru^{99} below 700 keV.

A number of experiments investigating the nature of the ground and first-excited states of Ru^{99} have been performed. The spin of the ground state has been measured as $\frac{5}{2}$ by Griffith and Owen.[5] Murakawa[6] has found the magnetic moment of the ground state to be -0.63 nm. Using the Mössbauer effect to study the hyperfine structure of the first-excited state (89.4 keV), Kistner[7] has reported that the spin of this level is $\frac{3}{2}$, and that the mixing ratio of 89.4-keV γ ray is $\delta^2=2.7\pm0.6$. The lifetime of the 89.4-keV level has been measured by Kistner, Monaro, and Schwarzschild[8] and by Matthias, Rosenblum, and Shirley[9] with the latter group reporting that $T_{1/2}$(89.4-keV level)$=20.7\pm0.3$ nsec. Kistner[7] and Matthias[9] *et al.* have presented measurements of the *g* factor of the first-excited state. Kistner's experiment,[7] using Mössbauer techniques, gave the ratio of the *g*

† Work supported by the U. S. Atomic Energy Commission.
* Present address: Physics Department, University of Manitoba, Winnipeg, Manitoba, Canada.

[1] J. D. Kurbatov, C. W. Townley, B. Feigley, and M. H. Kurbatov, Bull. Am. Phys. Soc. **5**, 240 (1960).

[2] S. Jha, S. Johnston, and T. D. Nainan, Bull. Am. Phys. Soc. **8**, 524 (1963).

[3] *Nuclear Data Sheets*, compiled by K. Way *et al.* (Printing and Publishing Office, National Academy of Sciences—National Research Council, Washington 25, D. C.), NRC-61-1-50.

[4] P. I. Connors, thesis, Pennsylvania State University, 1966 (unpublished).

[5] J. H. E. Griffiths and J. Owen, Proc. Phys. Soc. (London) **A65**, 951 (1952).

[6] K. Murakawa, J. Phys. Soc. Japan **10**, 919 (1955).

[7] O. C. Kistner, Phys. Rev. **144**, 1022 (1966).

[8] O. C. Kistner, S. Monaro, and A. Schwarzschild, Phys. Rev. **137**, B23 (1965).

[9] E. Matthias, S. S. Rosenblum, and D. A. Shirley, Phys. Rev. **139**, B532 (1965).

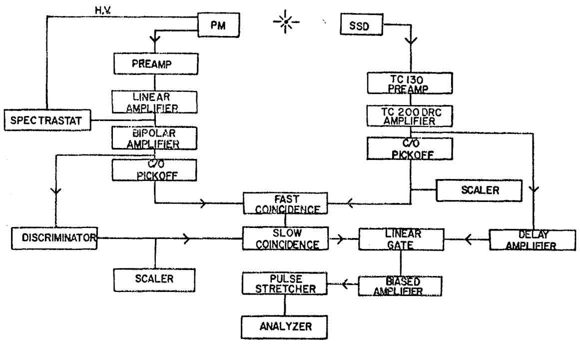

FIG. 1. A block diagram of the electronics used for the coincidence and directional correlation studies.

factors of the 89.4-keV level and the ground state as 0.759 ± 0.016 so that $g(89.4\text{ keV})=-0.19\pm0.05$. Matthias[9] *et al.*, taking advantage of the long halflife of the first-excited state, measured the g factor of this level by investigating the rotation of the angular correlation pattern in an external magnetic field. The results were $g(89.4\text{ keV})=-0.189\pm0.004$, leading to a magnetic moment for this state of -0.284 ± 0.006 nm.

Directional correlation measurements on the 353.0–89.4- and 527.7–89.4-keV cascades have been reported by Leonard and Jha.[10] They obtained[11] for these cascades the coefficients $A_2=-0.20$ and $A_2=-0.082$, respectively, where neither of these results are corrected for solid angle. Matthias[9] *et al.* repeated the measurements to verify the anisotropies of these correlations and found the anisotropy for the 353.0–89.4-keV correlation to be $A=-(15\pm2)\%$ while for the 527.7–89.4-keV cascade $A=-(19\pm2)\%$. No correction for solid angle or background was made. Connors[4] has observed the directional correlations arising from the (353.0–89.4)-, (527.7–89.4),- and (175.2–353.0)-keV cascades and has found the coefficients $A_2=-0.135$, $A_2=-0.13$, and $A_2=+0.22$, respectively.

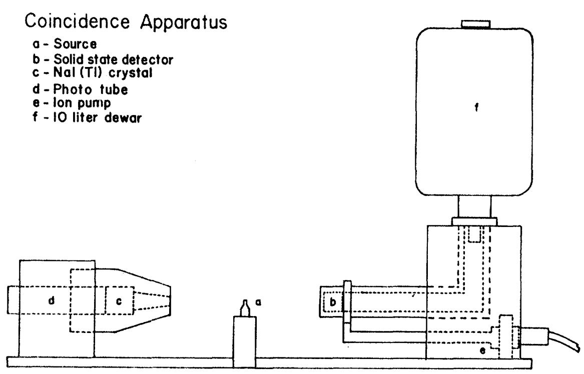

FIG. 2. The source and detector arrangement of the correlation table.

[10] R. F. Leonard and S. Jha, Bull. Am. Phys. Soc. **9**, 484 (1964).
[11] These are the values reported by the authors at the conference cited, not those found in the abstract (see Ref. 9).

TABLE I. γ-ray calibration sources.

Isotope	γ-ray energies (keV)	Reference
I^{131}	80.164±0.009	a
	284.307±0.049	a
	364.467±0.050	a
Na^{22}	511.006±0.002	b
Y^{88}	898.0 ±0.3	c
Bi^{207}	1063.51 ±0.08	d
Na^{22}	1274.7 ±0.2	d
Co^{56}	1359.9 ±0.3	c
	1770.8 ±0.4	c
Y^{88}	1836.2 ±0.3	d
Co^{56}	2015.6 ±0.7	c
	2034.7 ±0.3	c
	2598.9 ±0.3	c
	3202.3 ±0.5	c
	3254.0 ±0.5	c

[a] H. C. Hoyt and J. W. M. DuMond, Phys. Rev. **91**, 1027 (1953).
[b] E. R. Cohen and J. W. M. DuMond, Rev. Mod. Phys. **37**, 537 (1965).
[c] K. W. Dolan, D. K. McDaniels, and D. O. Wells, Phys. Rev. **148**, 1151 (1966).
[d] J. B. Marion (private communication, 1966).

$B(E2)$ values for various excited states in Ru^{99} have been obtained from the Coulomb excitation experiments of Temmer and Heydenburg[12] and more recently by Kistner and Schwarzschild.[13] The enhancements of the measured $B(E2)$ values over the single-particle estimates are indicative of the collective nature of some of the states.

Because of the complexity of the γ-ray spectrum, the experimental results which led to the decay scheme found in the Nuclear Data Sheets are of questionable validity. The purpose of the present investigation is to make use of the excellent energy resolution of lithium-drifted germanium solid-state detectors [Ge(Li) detectors] to study the γ-ray spectrum of Ru^{99} resulting from the decay of the 16.1-day isomer of Rh^{99}. Ge(Li) detectors have been utilized to establish accurate γ-ray energies and transition intensities. A decay scheme has been formulated with the aid of coincidence measurements. The spins of some of the levels have been investigated through directional correlation measurements using a Ge(Li) detector in conjunction with a NaI(Tl) spectrometer.

In Sec. II, the experimental apparatus and procedures used in this investigation are discussed. The analysis of the observed γ-ray spectrum and directional correlation measurements is presented in Sec. III. An attempt is made to describe the low-lying states of Ru^{99} using the intermediate coupling model of Bohr and Mottelson and the Nilsson model in Sec. IV. A brief comparison with the predictions of the core-excitation model of de-Shalit is also included.

[12] G. M. Temmer, and N. P. Heydenburg, Phys. Rev. **104**, 967 (1956).
[13] O. C. Kistner and A. Schwarzschild, Phys. Rev. **154**, 1182 (1967).

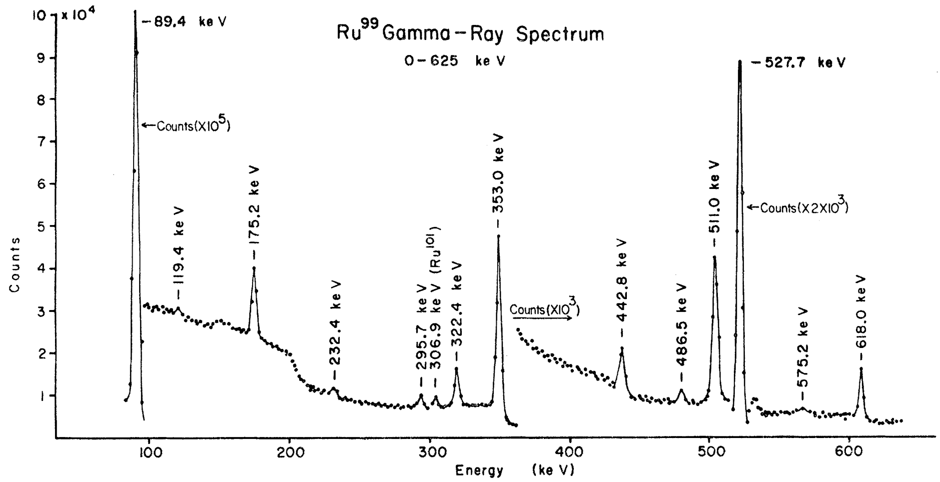

FIG. 3. The Ru^{99} γ-ray spectrum as observed with the Ge(Li) detector: 0–635 keV.

II. EXPERIMENTAL

A. Source Preparation

Ruthenium metal enriched to 98.26% Ru^{99} was obtained from the Oak Ridge National Laboratory Isotope Center. Since the metal was in an amorphous form, it was easily ground into a fine powder and packed between layers of pure aluminum foil 1 mil thick. The target package was bombarded with 10 MeV protons from the University of Washington cyclotron; the energy of the beam was degraded to 6.5 MeV by foils placed in front of the target. Two separate sources were prepared with total beam currents of 620 and 542 μA. The rhodium activity was produced by the $Ru^{99}(p,n)$-Rh^{99} reaction, which has a Q value of -2.883 MeV. The 6.5-MeV beam energy was chosen to avoid as many of the contaminating reactions as possible. No chemical separation was attempted.

B. Apparatus and Procedures

A Ge(Li) detector[14] with a sensitive area of 150 mm^2 and a depletion depth of 3 mm was used for energy measurements up to 1 MeV and for all relative intensity measurements. The cryostat and associated electronics has been described elsewhere.[15] The energy resolution of the system ranged from 3.2-keV full width at half maximum (FWHM) for the 661.59-keV Cs^{137} γ-ray to 4.0 keV for the 1173.23- and 1332.48-keV Co^{60} γ-rays. To confirm energy measurements made with the 3-mm detector and for the analysis of the spectrum above 1 MeV, a 6-cm^3 axially-drifted Ge(Li) detector[16] was employed. Through the use of a field-effect transistor preamplifier, the energy resolution of this detector system was 4.2-keV FWHM for the Co^{60} γ rays.

The energy of each γ-ray was determined by performing a least-squares fit to the expression $E=a+bx+cx^2$, where E is the energy and x is the channel number of the centroid of the photopeak whose energy is to be evaluated. The calibration sources used to determine the constants in this expression are given in Table I. The relative intensity measurements were performed using the 3-mm Ge(Li) detector and the photopeak efficiency calculations made by Dolan *et al.*[17]

Coincidence investigations and $\gamma-\gamma$ directional correlations were performed with the 6-cm^3 Ge(Li) detector in conjunction with a gain stabilized NaI(Tl) spectrometer.[18] A block diagram of the coincidence configuration is shown in Fig. 1. The resolving time of the fast coincidence circuit was 40 nsec. These measurements were performed on a correlation table which was constructed to allow very accurate centering of the source with respect to the detectors. A cylindrical Lucite cup 2 mm deep and 1 mm in diameter with a wall less than 0.5 mm thick was used as the source holder. The scintillation detector was shielded with a precisely machined lead cone which defined a half angle of 5.1°. A diagram of this apparatus is shown in Fig. 2.

III. ANALYSIS AND RESULTS

A. γ-Ray Energies and Intensities

The results of the γ-ray singles measurements are shown in Figs. 3–6; these spectra are typical of a large number of observations made with the Ge(Li) detectors. Figure 3 illustrates the energy region from 80 to 625 keV. The line at 306.9 keV is due to Ru^{101} and was observed to decay rapidly enough to distinguish it from the surrounding γ rays. Connors[4] has stated that the 575.2-keV transition is populated by the decay of the 4.3-h isomer of Rh^{99}; from our observations, no differ-

[14] Obtained from Solid State Radiations, Los Angeles 69, California.

[15] P. V. Rao, D. K. McDaniels, and Bernd Crasemann, Nucl. Phys. **81**, 296 (1966).

[16] Obtained from Princeton Gamma-Tech, Princeton, New Jersey.

[17] K. W. Dolan, D. K. McDaniels, and D. O. Wells, Phys. Rev. **148**, 1151 (1966).

[18] Harshaw Integral Line Assembly, Harshaw Chemical Co., Cleveland, Ohio. The crystal size was 1.5 in.×1.75 in.

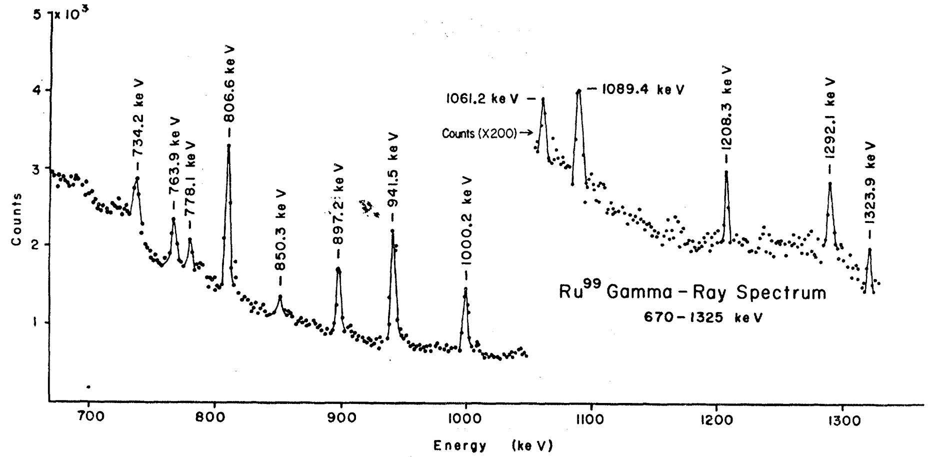

FIG. 4. The Ru^{99} γ-ray spectrum as observed with the Ge(Li) detector: 670–1325 keV.

ence between the decay rate of this line and neighbor transitions could be observed. Therefore, it has been included in the 16.1-day Rh^{99} decay.

The γ-ray spectrum from 670 to 1325 keV is shown in Fig. 4. The γ ray at 778.1 keV does not belong to the 16.1-day decay; the origin of this line has not been explicitly determined.

Figure 5 presents the spectrum in the region between 1350 and 1760 keV. The lower efficiency of the detector and the inherent weakness of the high-energy transitions necessitated longer data-collection times. Although the energy spectrum below 1 MeV could be obtained in periods from 1–4 h., the data collected above 1 MeV required observation intervals of 8–24 h. The effect of the longer running times affected the energy resolution of the system by less than 1.5 keV FWHM.

The transitions in Ru^{99} above 1900 keV are shown in Fig. 6. Co^{56} lines are included in this spectrum as calibration standards for this energy region.

Although they are not all evident in the spectra shown, the presence of other ruthenium isotopes was noted: The 539.4-keV line of Ru^{100}, the 306.9-keV line of Ru^{101}, and the 474.5-keV transition in Ru^{102} were the only measureable contributions. Several γ rays originating from the 4.3 h isomeric state in Rh^{99} were also seen. A listing of the γ rays observed in Ru^{99} and their intensities relative to that of the 89.4-keV transition is given in Table II.

B. Decay Scheme

To formulate the decay scheme of a nucleus with such a large number of γ-ray transitions (35+annihilation radiation) is a sizable task, and without the results of some coincidence measurements is virtually impossible. The weak intensity of the majority of the transitions and the relatively low γ-ray efficiency of the Ge(Li) detectors for the higher-energy γ rays would not permit as complete an investigation of the $\gamma-\gamma$ coincidences as desired. Figure 7 is an example of the coincidence spectra obtained with this system and indicates γ rays coincident with the 89.4-keV transition. Table III summarizes the results of these measurements.

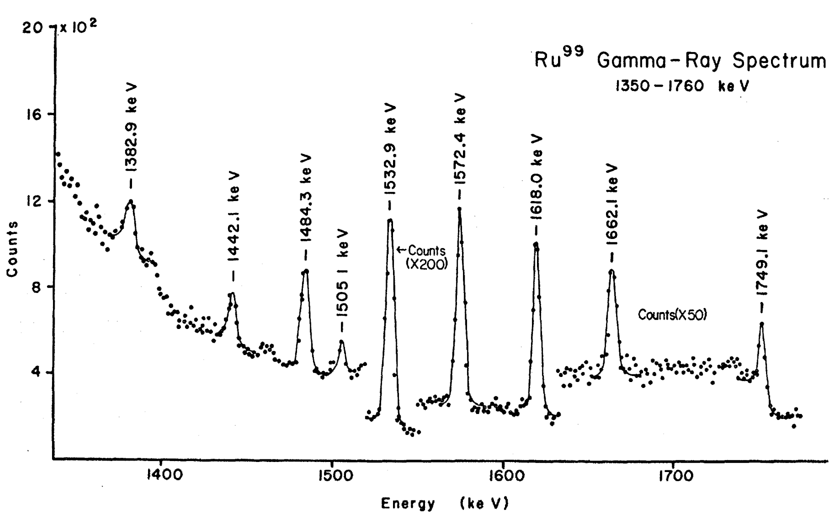

FIG. 5. The Ru^{99} γ-ray spectrum as observed with the Ge(Li) detector: 1350–1760 keV.

TABLE II. γ-rays observed in the decay of 16.1-day Rh^{99}

Energy[a] (keV)	Relative intensity
89.36	100
119.4	0.17
175.2	7.6
232.4	1.7
295.7	2.8
322.4	21.7
353.0	107.1
442.8	5.7
486.5	2.0
annihilation radiation	
527.7	132.0
575.2	0.8
618.0	13.1
734.2	1.1
763.9	1.3
806.6	4.6
850.3	0.8
897.2	2.2
941.5	5.0
1000.2	2.3
1061.2	0.6
1089.4	1.0
1208.3	0.6
1292.1	1.2
1323.9	0.5
1382.9	0.6
1442.1	0.3
1484.3	0.5
1505.1	0.3
1532.9	1.8
1572.4	7.7
1618.0	0.8
1662.1	0.2
1749.1	0.2
1970.3	0.5
2058.6	0.1

[a] Accuracy of energies is ±0.4 keV or better for transitions less than 1 MeV, and ±0.7 keV for the 1- to 2-MeV range.

To aid in building a decay scheme, a program was written for the IBM 360/50 computer which formed all possible sums of two, three, and four γ rays (the sums retained were those whose energy was less than the mass difference[19] between Rh^{99} and Ru^{99}).[20] The program then searched the γ-ray spectrum for transitions equal to any of the sums (within a prescribed test criterion); this selected all possible cascade transitions for a given crossover transition. In addition, the program formed all possible sums of two γ rays to determine whether any common sums existed and to check whether there might be a level in the decay scheme which had no crossover transition, but which did have two or more γ-ray cascades originating from it. With this information and the coincidence spectra, the decay scheme shown in Fig. 8 was constructed.

The level structure below 618.0 keV was easily determined, since strong γ-ray coincidences were observed for most of the transitions involved. This part of the decay scheme is in agreement with the observations made by Connors[4] on the low-lying states in Ru^{99} with the exception of the 575.2-keV level which he states is populated by the 4.3-h isomeric state only. We have observed this state to be fed by the 16.1-day decay and have included it in this decay scheme. The search program found that the 119.4-keV γ ray could have fit into several locations in the scheme. However, the coincidence spectra obtained from gating on the 322.4-keV transition indicated that the 119.4-keV line was coincident with the 322.4-keV γ ray.

The 763.9-keV transition could be placed in coincidence with the 527.7-keV γ ray, but then should be observed coincident with this very strong line; it is not. Furthermore, since the sum[21] of these transitions equals 1292.1 keV, the crossover transition would necessarily be in coincidence with the 89.4-keV ground-state transition. However, there was no indication of such a coincidence. The remaining possible location of the 763.9-keV γ ray as presented by the search program is shown on the decay scheme.

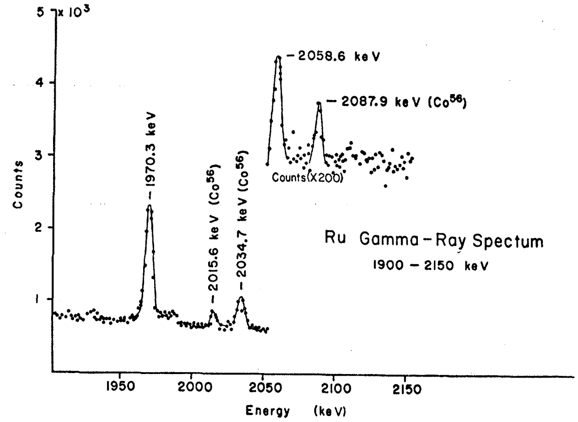

FIG. 6. The Ru^{99} γ-ray spectrum as observed with the Ge(Li) detector: above 1900 keV. The Co^{65} lines are for energy calibration.

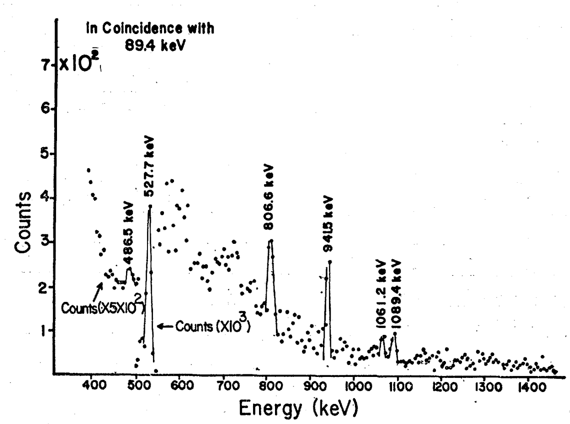

FIG. 7. An example of the coincidence spectra obtained. γ-rays in the energy region 400–1400 keV that are found in coincidence with the 89.4-keV transition.

[19] The total decay energy has been measured and found to be 2.11±0.025 MeV (Ref. 4).

[20] J. H. E. Mattauch, W. Thiele, and A. H. Wapstra, Nucl. Phys. **67**, 1 (1965).

[21] In this discussion the sum of two transitions which "equals" a third implies that the sum is equal to the third within the test criterion given to the search program (1 keV in most instances).

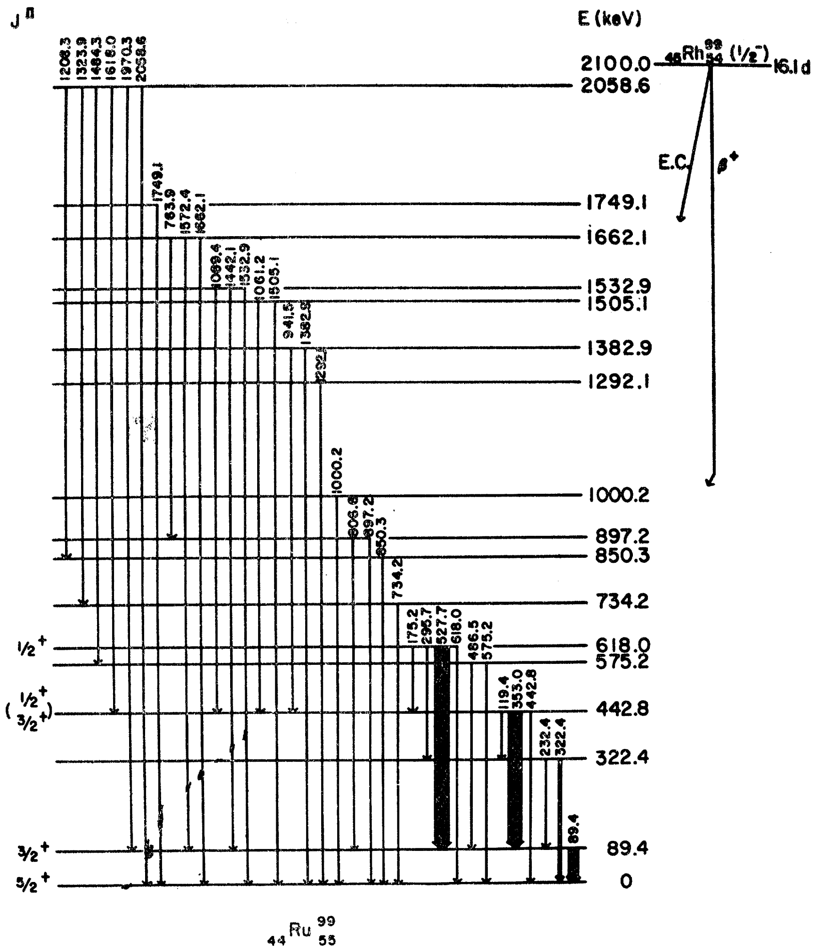

FIG. 8. The energy-level diagram of the excited states of Ru^{99} populated by the decay of 16.1-day Rh^{99}.

A potential position for the 850.3-keV γ ray is in cascade with the 89.4- and 353.0-keV transitions, but the 850.3-keV γ was not observed in coincidence with the strong 353.0-keV line. The cascade formed by the 850.3- and the 1208.3-keV γ's is the only suitable location for the latter transition that was found by the search routine. It is conceivable that the order of these γ rays could be reversed in the cascade.

There are three apparent locations for the 1000.2-keV line in the decay scheme. Careful observations were made to find either an 89.4–1000.2,- 353.0–1000.2-, or 527.7–1000.2-keV coincidence. Each of these coincidences could be easily observed if they existed since the intensities of the contributing lines are all significantly large. None of the above combinations was found. Since the search program could find no other potential location for the 1000.2-keV transition, it is assumed a ground-state transition.

The spectrum of γ rays coincident with the 89.4-keV transition was checked for the presence of the 1292.1-keV γ since the search program found only combinations that involved the 89.4-keV line; no indications of the 1292.1-keV γ ray are found in the 89.4-keV coincidence spectrum.

The search code found that the 1442.1-keV γ ray could be located in two positions: terminating either on the 89.4- or the 618.0-keV level. The 527.7-keV coincidence spectrum showed no sign of any transitions other than the 89.4 keV; the 89.4 coincidence results revealed only a very few 1442.1-keV events. With this information, the 89.4–1442.1-keV cascade is considered the most likely. However, the possibility of a 618.0–1442.1-keV cascade cannot be ignored.

The remainder of the γ-rays were located in the decay scheme with no ambiguity. For the cascades involving transitions above 1 MeV, the test criterion of the search program was relaxed to 1.4 keV because of the somewhat reduced accuracy in determining the energy of the weaker high-energy transitions.

C. Directional Correlation Measurements

The data for the directional correlation measurements were taken at four angles: 90°, 120°, 150°, and 180° for the 89.4–527.7- and 89.4–353.0- keV cascades, and at 70°, 90°, 120°, and 150° for the 175.2–353.0-keV cascade. Data could not be taken at 180° for the latter cascade since the backscatter energy of the strong 527.7-keV γ ray is 172.2 keV. The NaI(Tl) spectrometer was always used as the "gate", making the energy selection, and the coincidence spectrum was observed with the Ge(Li) detector. In this way the excellent energy resolution of the solid-state detector could be used to resolve the coincident γ rays.

The 353.0-keV photopeak lies directly on top of the compton edges of the annihilation radiation and the 527.7-keV γ ray. Thus it was necessary to remove the contribution of these radiations from beneath the 353.0-keV photopeak. This was accomplished by constructing the single line shape of the 511-keV radiation using a Na^{22} source. The full-energy peak of this radiation was normalized to the full-energy peak of the γ ray, whose Compton distribution was to be subtracted and the contributions due to the unwanted radiation were removed.

Each observed directional correlation was fit to the expression

$$W(\theta)=\sum_k A_{2k}P_{2k}(\theta), \quad k=0,1,2, \tag{1}$$

by the method of least squares, where the $P_{2k}(\theta)$ are Legendre polynomials. Since the spin of the 89.4-keV level is known to be $\frac{3}{2}$, only the $k=0$ and 1 terms were used in the expansion when this level was the intermediate state in the observed cascade.

The measured directional correlations for the 89.4–353.0 and 89.4–527.7-keV cascades are shown in Fig. 9 and the solid line is the result of the least-squares fit to the expression (1). With the correction made for the finite geometry of the detectors, we find that

$$W(\theta)=1-(0.131\pm0.012)P_2(I)$$

for the 89.4–353.0-keV cascade. The corrected correla-

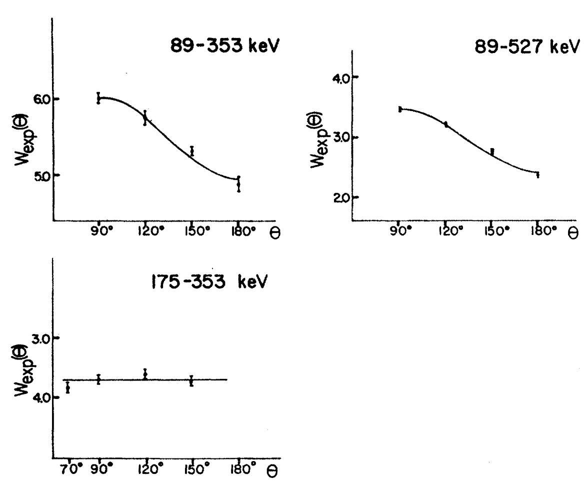

FIG. 9. The $\gamma-\gamma$ directional correlation for the (a) 89.4–353.0-keV cascade, (b) 89.4–527.7-keV cascade, and (c) 175.2–353.0-keV cascade. The solid line is the result of a least-squares fit to Eq. (1).

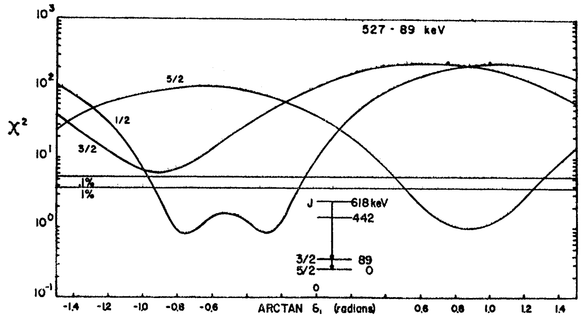

FIG. 10. χ^2 as a function of the arctangent of the mixing ratio of the 527.7-keV transition with j(618.0-keV level) $=\frac{1}{2}$, $\frac{3}{2}$, and $\frac{5}{2}$.

tion for the 89.4–527.7-keV cascade is

$$W(\theta)=1-(0.221\pm0.020)P_2(\theta).$$

Figure 9 also illustrates the observed 175.2–353.0-keV correlation, where

$$W(\theta)=1-(0.012\pm0.078)P_2(\theta)-(0.043\pm0.162)P_4(\theta).$$

This result is not in agreement with the measurement made by Connors.[4]

The analysis of the correlation results has been made using the χ^2 test. For these purposes the quantity χ^2 is defined as

$$\chi^2=\frac{1}{n}\sum_k [W(\theta_k,\delta)-Y(\theta_k)]^2/E^2(\theta_k),$$

where n is the number of degrees of freedom,[22] $W(\theta_k,\delta)$ is the theoretical correlation function which depends on the mixing ratio[23] of the γ rays in the cascade and on the spins of the levels, $Y(\theta_k)$ is the normalized yield at each angle θ_k, and $E(\theta_k)$ is the error associated with the yield measurement.

Figure 10 is a plot of χ^2 versus the arctangent of the mixing ratio of the 527.7-keV transition for various spins of the 618.0-keV level. The measured value of the mixing ratio of the 89.4-keV γ ray[7] has been used in the calculation. The 0.1% and 1% limits that are given by the χ^2 probability tables are indicated in the figure. It can be seen that both the $\frac{1}{2}$- and $\frac{5}{2}$-spin assignments for the 618.0-keV level are consistent with the measurements. The logft value of the 618.0-keV level has been calculated to be 7.0; this leads to $j(618.0\text{ keV})=\frac{1}{2}$ or $\frac{3}{2}$ (unique first-forbidden decay). Thus the $j=\frac{5}{2}$ solution suggested by the χ^2 plot can be eliminated, and the spin of the 618.0-keV level is $\frac{1}{2}$. The range of the mixing ratio of the 527.7-keV transition (values for which χ^2 is less than the 1% limit) is $0.013\leq\delta^2\leq1.84$.

The χ^2 plot of the 353.0–89.4-keV correlation is given in Fig. 11. As can be seen from this illustration no unique decision can be made about the spin of the 442.8-keV

[22] The number of degrees of freedom is defined to be the number of observations k minus the number of adjustable parameters.

[23] The mixing ratio of a γ-ray transition is given by $\delta^2=E2/M1$, i.e., the ratio of the intensity of $E2$ radiation to the intensity of $M1$ radiation in the transition.

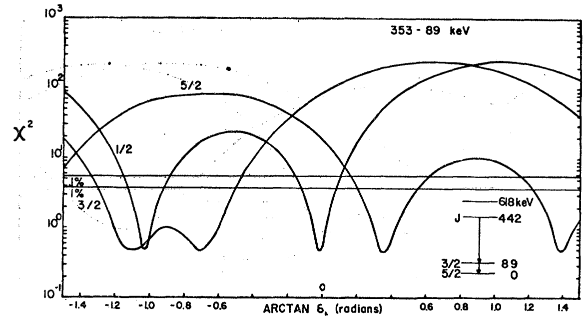

FIG. 11. χ^2 as a function of the arctangent of the mixing ratio of the 353.0-keV transition with j(442.8-keV level) $= \frac{1}{2}$, $\frac{3}{2}$, and $\frac{5}{2}$.

level. A logft value of 7.4 has been computed for the transition to this level. This corresponds to a first-forbidden decay and thus a spin assignment of either $\frac{1}{2}$ or $\frac{3}{2}$ for the 442.8-keV level.

The analysis of the 175.2–353.0-keV correlation is shown in Fig. 12. The least-squares fit to the data reveals that both the A_2 and A_4 coefficients are equal to zero within the experimental errors. This isotropy suggests that the spin of the 442.8-keV level might be $\frac{1}{2}$. Figure 12 indicates, however, that the $j=\frac{3}{2}$ assignment cannot be discounted. The K/L ratio measurement for the 353.0-keV transition[4] is not known to sufficient accuracy to assist in the determination of the spin of the 442.8-keV level. Hence, we can only conclude from the above measurements that the spin of the 442.8-keV level is either $\frac{1}{2}$ or $\frac{3}{2}$, with the more likely assignment being $\frac{1}{2}$.

IV. DISCUSSION

The experimental data from the studies of the excited states of Ru^{99} suggest a collective description for this nucleus. For example, the $E2$ transition rate[8] for the first-excited state (89.4 keV) is 50 times the single-particle estimate and the $M1$ rate is retarded by about 2×10^4. Further information is provided by data from a tabulation of $B(E2)$ values due to Stelson and Grodzins.[24] They have plotted the ratio β_2/β_{2SP} for even-even nuclei as a function of neutron number.[25] Figure 13 illustrates the behavior of this quantity for the even ruthenium isotopes; for comparison the values of β_2/β_{2SP} for several other nuclei are also indicated on the figure. It is clear from this diagram that the nuclear deformation is increasing with mass number in this region. We have used three unified nuclear models to attempt to describe the observed energy-level spectrum of Ru^{99}.

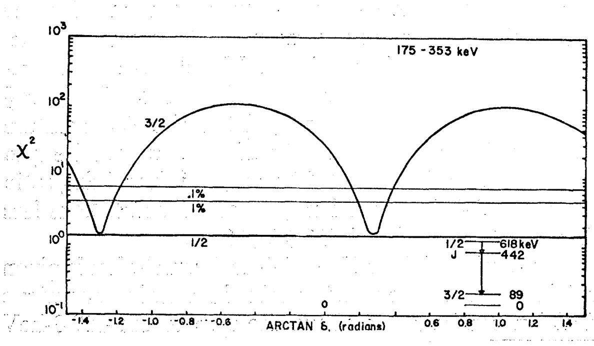

FIG. 12. χ^2 as a function of the arctangent of the mixing ratio of the 175.0-keV transition with j(442.8-keV level) $=\frac{1}{2}$ and $\frac{3}{2}$. The spin of the 618.0-keV state is taken to be $\frac{1}{2}$.

[24] P. H. Stelson and L. Grodzins, Nucl. Data **1A**, 21 (1965).

A. Intermediate Coupling Model

The coupling of single-particle states to a collective nuclear core has been treated extensively by Bohr and Mottelson,[26] where various coupling strengths between the particle and core states were examined. With the intermediate coupling, one considers the interaction between collective surface vibrations of the even-even core and a single extra-core nucleon which has several single-particle states available. The resulting energy-level spectrum will depend on the single-particle spacing, the magnitude of the surface vibrations, the coupling strength of the particle-core system, and the final spin of the coupled system. This calculation has been treated in detail elsewhere[26–28] and will not be repeated here.

Figure 14 illustrates the results of a one-phonon calculation where only the lowest few states are shown. The energy levels are plotted as a function of the parameter ξ which determines the strength of the core-particle coupling. The one-phonon excitation energy used was the energy of the first-excited state of the Ru^{98} core, 654 keV. The single-particle states available to the neutron are the $1g_{7/2}$, $3s_{1/2}$, and the $2d_{3/2}$; the splittings chosen were $g_{7/2}-d_{5/2}=1.5$ MeV, $s_{1/2}-d_{5/2}=1.2$ MeV, and $d_{3/2}-d_{5/2}=1.4$ MeV. It can be seen from the figure that the interaction does not successfully predict either the proper spin or energy of the first-excited state.

A further possibility is to assume that the energy-level spectrum can be explained by coupling a neutron hole to the Ru^{100} core. If it is assumed that the hole-core interaction is the same as the particle-core interaction, with the exception of a change in sign, it can be seen that the states that were pushed up in energy as a result of the particle-core interaction would be brought down in the case of a hole-core interaction, while those states lowered in the former case would be increased in energy in the latter situation. From Fig. 14 one can determine what is likely to occur: the $\frac{5}{2}$ state, which is repelled from the center of gravity of the multiplet by the particle-core

[25] The nuclear deformation parameter β_2 is given by $\beta_2=[B(E2), 0\rightarrow 2]^{1/2}/(3ZR_0^2/4\pi)$, where R_0 is the nuclear radius. The single-particle deformation parameter β_{2SP} is given by the same expression, except that the reduced transition probability is $B(E2)_{SP}=2.95\times10^{-53}A^{4/3}$.

[26] A. Bohr and B. R. Mottelson, Kgl. Danske Videnskab. Selskab, Mat. Fys. Medd. **27**, 16 (1957).

[27] D. C. Choudhury, Kgl. Danske Videnskab. Selskab, Mat. Fys. Medd. **28**, 4 (1954).

[28] N. K. Glendenning, Phys. Rev. **119**, 213 (1960).

TABLE III. A summary of the coincidence measurements.

E (keV)	In coincidence with E
89.4	175.2,[a] 232.4, 295.7, 353.0, 486.5, 527.7, 806.6, 941.5,[a] 1061.2,[a] and 1089.4[a] keV.
322.4–353.0	89.4, 119.4, 295.7, 941.5, 1061.2, and 1089.4 keV.
527.7	89.4 keV

[a] In coincidence with the 89.4-keV transition through the 353.0-keV γ ray.

interaction, would instead be attracted and would become the first-excited state, thereby failing to reproduce the observed spin sequence. Thus, one can conclude that this nucleus is not properly described by the coupling of a single nucleon to a vibrational core with the interaction specified by this model.[28] Perhaps the particle-core coupling is relatively strong, implying a deformed nucleus, and the approximations made in this type of calculation are not valid.

B. Nilsson Model

In regions far from closed shells the nucleus assumes a deformed shape and it becomes possible to distinguish between the intrinsic particle motion relative to the deformed nuclear field and the collective excitations of the core. The theoretical description of single-particle motion in a deformed nuclear potential was first developed by Nilsson.[29] The interaction between the nucleon and the nuclear field is represented by a spheroidal harmonic-oscillator potential with spin-orbit interaction and a term proportional to the square of the orbital angular momentum. The Hamiltonian is

$$H=H_0+C\mathbf{l}\cdot\mathbf{s}+Dl^2,$$

where H_0 is the spheroidal oscillator term. The parameters C and D are not completely free to choose since in the limit of spherical symmetry the level ordering should be identical with the shell-model predictions. The Hamiltonian is rearranged and expressed in terms of the parameters κ, μ, and ϵ, where κ and μ are directly proportional to C and D, respectively, and where ϵ is the nuclear deformation parameter.[29] The resulting matrix is diagonalized and the eigenvalues are calculated for various values of ϵ, μ, and κ, subject to the restriction that as $\epsilon\rightarrow 0$ the single-particle level ordering should be preserved. For the $N=4$ shell, Nilsson chose $\mu=0.45-0.55$, which reproduced the shell-model sequence of states as given by Klinkenberg.[30] The influence of the value of μ on the level ordering for zero deformation ($\epsilon=0$) is shown in Fig. 15. Recently Cohen[31] has published a compilation of current experimental data from which the neutron single-particle spacings have been deduced over a wide range of the periodic table. The ordering used by Nilsson is found for values of μ in the range 0.45–0.55, while the single-particle sequence expected from Cohen's presentation is found for $\mu=0.1$–0.2. Figures. 16 and 17 are the Nilsson diagrams for the $N=4$ shell, with $\kappa=0.125$ and $\mu=0.1$ and 0.2. For positive deformations up to $\epsilon\sim 0.2$, the $\frac{5}{2}$ ground state is successfully predicted; the ground-state magnetic moment calculated[32] for $\mu=0.1$ and $\epsilon=0.2$ is -0.96 nm, which is comparable with the measured value of -0.63 nm. The $\frac{3}{2}$ first-excited state present in the energy-level spectrum must be an intrinsic state and should be predicted by the model. Since no reasonable combination of parameters would predict a first-excited state with spin $\frac{3}{2}$, we must conclude that this model also is not an adequate representation of this nucleus.

[29] S. G. Nilsson, Kgl. Danske Videnskab. Selskab, Mat. Fys. Medd. **29**, No. 16 (1960).
[30] P. Klinkenberg, Rev. Mod. Phys. **24**, 63 (1952).
[31] B. L. Cohen, Am. J. Phys. **33**, 1011 (1965).

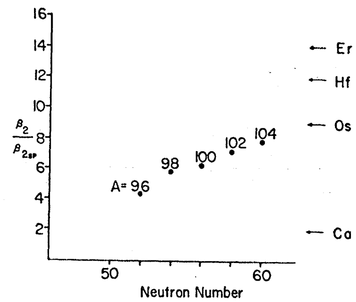

FIG. 13. The ratio β_2/β_{2SP} as a function of neutron number for the even ruthenium isotopes (Ref. 15). The value of this ratio for various other nuclei is indicated for comparison.

C. Core-excitation Model

If the single-particle states of the odd nucleon in an odd-A nucleus are separated from the ground state by

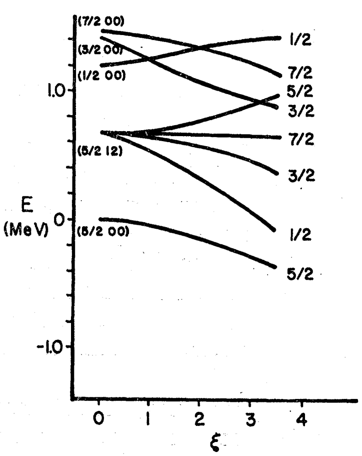

FIG. 14. The energy-level spectrum as a function of the strength parameter ξ (see text) resulting from the intermediate-coupling-model calculation. The numbers in the parentheses are the quantum numbers (j,N,R) where j is the angular momentum of the single-particle level, N is the number of surface phonons, and R is the angular momentum of the surface.

[32] For this calculation g_R was taken to be equal to Z/A.

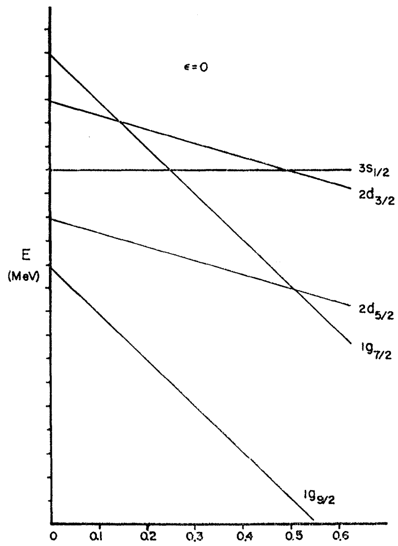

FIG. 15. The effect of the l^2-strength parameter μ in the Nilsson Hamiltonian on the single-particle spacings (for zero deformation).

relatively large energies, it is reasonable to assume that the low-lying states of the odd nucleus can be described by including excitations of the nuclear core. This is the basis of the intermediate coupling scheme presented earlier, where the core excitations are assumed to be vibrational modes described by a harmonic oscillator Hamiltonian. de-Shalit[33] has suggested a version of this model which avoids any identification of the nature of the core excitations, and attempts to indicate the effects of coupling the core states to the odd nucleon in a completely general way. A set of states corresponding to the coupling of a single nucleon to the 2^+ state of the even-even core is considered to be the description of the lowest states in the neighboring odd-A nucleus. The ground state is the result of the odd nucleon being in the lowest state provided by the potential due to the even core. With the single-particle states removed in energy, the lowest levels in the odd nucleus are described by the coupling of the single nucleon in its ground state to excitations of the even core. Using an arbitrary interaction, de-Shalit has extended this calculation to include expressions for the $E2$ and $M1$ transition rates between members of the core-excitation multiplet and has derived the center-of-gravity theorem of Lawson and Uretsky.[34]

A discussion of the application of this model to Ru^{99} (as well as Ru^{101}) has been given by Kistner and Schwarzschild.[13] They point out that the model predicts a multiplet formed by the coupling of the $2d_{5/2}$ neutron to the first 2^+ state in the even core (Ru^{98}). Thus the lowest excited states will have spins of $\frac{1}{2}$, $\frac{3}{2}$, $\frac{5}{2}$, $\frac{7}{2}$, and

[33] A. de-Shalit, Phys. Rev. **122**, 1530 (1961).

[34] R. D. Lawson and J. L. Uretsky, Phys. Rev. **108**, 1300 (1957).

TABLE IV. The ratio of excitation transition probabilities in Ru^{99} and Ru^{98}.[a]

Transition (keV)	y	$R=\frac{B(E2\downarrow)_{Ru^{99}}}{B(E2\downarrow)_{Ru^{98}}}$
89.4	$\frac{3}{2}$	1.4
322.4	$(\frac{5}{2},\frac{7}{2})$	0.02
340.4[b]	$(\frac{7}{2},\frac{9}{2})$	0.01
442.8	$(\frac{1}{2},\frac{3}{2})$	(0.4,0.2)
617.3[b]	$\frac{7}{2}$	(0.65)
719.2[b]	$(\frac{7}{2},\frac{9}{2})$	(1.0,0.8)

[a] From Ref. 13.
[b] These levels are observed in Coluomb excitation.

$\frac{9}{2}$ and the center of gravity of this multiplet should be equal in energy to the energy of the core state. Moreover, each of the members of the multiplet should have a $B(E2\downarrow)$ value[35] to the ground state, which is equal to the $B(E2\downarrow)$ of the $2^+ \rightarrow 0^+$ transition of the core (for Ru^{98}, $B(E2\downarrow) = 0.095e^2 \times 10^{-48}$ cm^4).

From the results of their Coulomb excitation experiments, Kistner and Schwarzschild calculated $B(E2\downarrow)$ values for the low-lying states in Ru^{99} using the relation

$$B(E2\downarrow) = ([2j_g+1)/(2j_e+1)]B(E2\uparrow),$$

where j_g and j_e are the spins of the ground state and excited state, respectively. The ratio R of the $B(E2\downarrow)$ values for these transitions in Ru^{99} to the $B(E2\downarrow)$ value for the $2^+ \rightarrow 0^+$ ground state transition in Ru^{98} is given in Table IV. The value of R is seen to be approximately unity for only three of the low-lying levels[36]: the 89.4-, 617.3-, and 719.2-keV states.

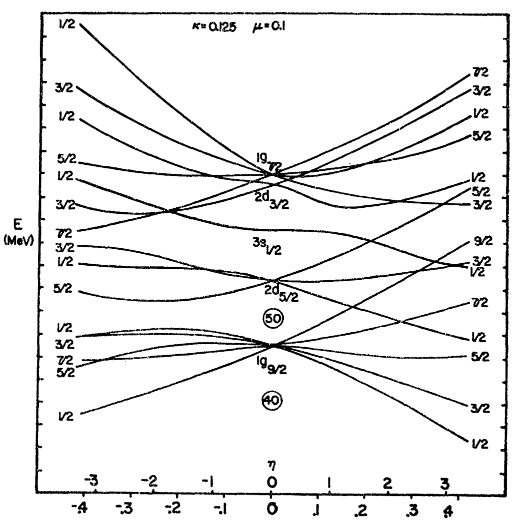

FIG. 16. The Nilsson-model energy-level spectrum as a function of the nuclear deformation for the $N=4$ shell; $\kappa=0.125$, $\mu=0.1$.

[35] The $B(E2\downarrow)$ value is the reduced transition probability for de-excitation from an excited state to the ground state.

[36] The 617.3- and 719.2-keV levels are not found in the decay of 16-day Rh^{99} but are seen in Coulomb excitation studies.

In attempting to find a series of levels which could represent the "core-excitation" multiplet, we recall that the multiplet will have five members (with spins $\frac{1}{2}$, $\frac{3}{2}$, $\frac{5}{2}$, $\frac{7}{2}$, and $\frac{9}{2}$). The 89.4-keV level is the obvious choice for the $j=\frac{3}{2}$ member of the multiplet. If the 442.8-keV level has $j=\frac{1}{2}$, then the value of R is found to be 0.4 as compared to 0.2 when $j=\frac{3}{2}$. Since this brings the ratio somewhat nearer to unity, we assume this to be the spin and include this level as the $j=\frac{1}{2}$ member of the multiplet. The $j=\frac{7}{2}$ member is taken to be the 617.3-keV state, while the 719.2-keV level is assumed to be the $\frac{9}{2}$ member. The $\frac{5}{2}$ member has yet to be determined. Table IV indicates only one level (322.4 keV) which is potentially spin $\frac{5}{2}$; the value of R associated with this level, however, is significantly different from unity. In attempting to find a $j=\frac{5}{2}$ level at higher energies, one must consider that Kistner and Schwarzschild reported that no transitions below 1.1 MeV were found which have a $B(E2\uparrow)$ from the ground state greater than $0.04e^2\times10^{-48}$ cm^4 other than those given in Table IV. If we assume that this is the value for the $j=\frac{5}{2}$ member of the multiplet, then R is found to be less than or equal to 0.4.

It remains to be determined at what energy the $j=\frac{5}{2}$ member would be found. de-Shalit has shown that the shift in energy of the multiplet states from the unperturbed energy can be expressed to first order by $\sum(2j+1)(E-E_c)=0$, where j is the spin of the level in the multiplet, E is the energy of the level with spin j, and E_c is the unperturbed energy. Thus the center of gravity of the multiplet is identical with its unperturbed position. Taking the $j=\frac{5}{2}$ member as the 1000.2-keV level we find that $E_c=645$ keV. This agrees favorably with the energy of the first-excited states in Ru^{98}, which is 654 keV. Figure 18 summarizes these results.

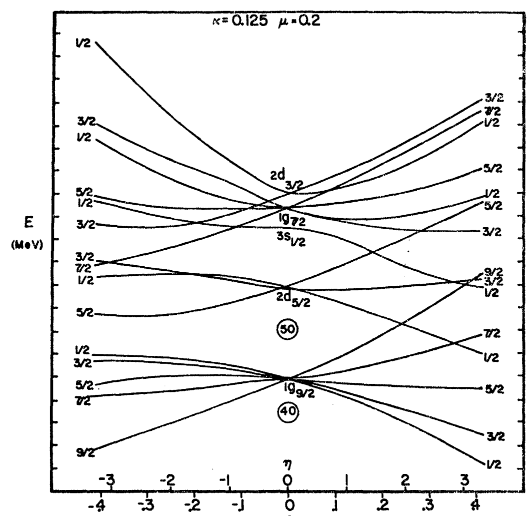

FIG. 17. The Nilsson-model energy-level spectrum as a function of the nuclear deformation for the $N=4$ shell; $\kappa=0.125$, $\mu=0.2$.

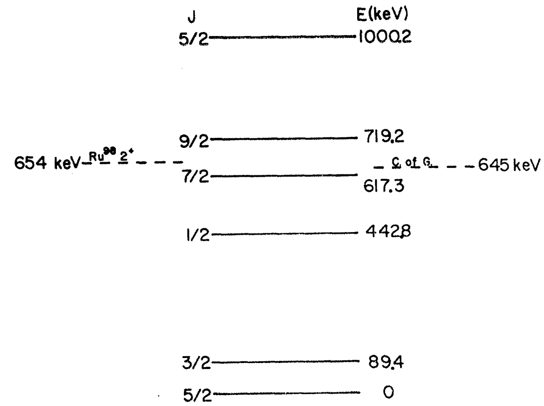

FIG. 18. A core-excitation multiplet resulting from the coupling of a single $\frac{5}{2}$ neutron to the 2^+ first-excited state in Ru^{98}. The center of gravity of the multiplet is shown in comparison to the energy of the first-excited state in Ru^{98}.

This discussion is not meant to suggest that a unique core-excitation multiplet has been found and is intended mainly as a heuristic device. Although some of the predictions of the model have been experimentally observed (such as fast $M1$ transitions between the levels which are members of the multiplet),[13] a number of questions remain unanswered. For example, there are several levels below 1 MeV which are not predicted by the model. Furthermore, the splitting between the members of the multiplet is too large. Nevertheless, of the three theoretical descriptions given above, the core-excitation model would seem to be the most consistent with the available experimental data. However, it is certainly apparent that the region around $A=100$ is not well represented by present-day nuclear-structure models.

ACKNOWLEDGMENTS

Discussions with Dr. D. O. Wells and Dr. M. K. Pal have been very helpful. Thanks are due to Dr. A. Schwarzschild for a preprint of the Coulomb excitation experimental results he obtained for ruthenium 99. The help of Joanne Sauer and the cyclotron crew at the University of Washington in preparing and bombarding the sources is greatly appreciated. The assistance of Miss Po Yuk Chee and Miss Betty Liu in analyzing some of the data has been most helpful.