Rotational States in Tm^{169}

SVEN A. E. JOHANSSON
Department of Physics, University of Lund, Lund, Sweden
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The decay of Yb^{169} has been investigated by means of a coincidence scintillation spectrometer. Eleven gamma rays have been arranged in a level scheme with levels at 10, 120, 142, 318, 336, 382, and 476 kev. The multipolarity of the strongest gamma rays has been determined and spin values assigned to some of the levels. It is found that five of the levels form a rotational band with $K=\frac{1}{2}$. In this case the Bohr-Mottelson unified model predicts an anomalous rotational spectrum. The experimental spectrum agrees very well with the theory. The gamma-ray intensities are discussed and there is a good agreement with the unified model also in this respect.

INTRODUCTION

THE ytterbium isotope of mass 169 decays by electron capture to Tm^{169}. The half-life is 32 days. The radiation emitted in this decay has been studied by several investigators[1,2] and is fairly well known. No consistent decay scheme seems to have been established, however.

The level scheme of Tm^{169} is of interest for a special reason. This nucleus lies in a region where the Bohr-Mottelson unified model[3,4] should be applicable. The lowest levels are expected to form a rotational band. The spin of the ground state is found to be $\frac{1}{2}$.[5,6] In this case the unified model predicts an anomalous spacing of the rotational levels. A determination of the level scheme will thus provide a rather detailed test of this model.

In the present work the decay of Yb^{169} has been investigated by means of a coincidence scintillation spectrometer. The experimental results make it possible to set up a consistent level scheme for Tm^{169}. This scheme is compared with the predictions of the unified model.

MEASUREMENTS

Radioactive ytterbium was obtained by pile irradiation of ytterbium oxide. The irradiated material (supplied by Johnson, Matthey and Company, Ltd., London) was of very high purity and had been tested by spectrophotometric and spectrographic examination. Foreign elements other than the rare earths were estimated to amount to less than 0.004% of the product. No other rare earth elements could be detected and the upper limit for the most likely impurities was estimated to be 0.02%. The ytterbium oxide was irradiated for one month and the sample was allowed to age for one month to allow the short-lived ytterbium activities to decay.

The coincidence scintillation spectrometer has been described in detail elsewhere.[7,8] It consists of two scintillation spectrometers coupled in coincidence. A certain radiation component is selected in the spectrum from one of the spectrometers and the corresponding coincidence spectrum from the other spectrometer is recorded. The resolving time of the coincidence circuit is 0.2 μsec. The scintillation spectrometer consists of sodium iodide crystals mounted on DuMont photomultipliers, type 6292. The resolution of the spectrometers is 6.2% full width at half-maximum for the Cs^{137} 662-kev line.

Figure 1 shows the gamma spectrum of Yb^{169}. The statistical errors in this spectrum as well as in all the following are small and have not been drawn in the figures. Most spectra comprise more than 10 000 pulses. The resolution of the spectrum in its main components is also shown. The highest peak is due to the K_α and K_β x-rays of thulium. They are emitted in the electron capture process and in the internal conversion of the gamma rays. The spectrum also contains gamma rays of the energies 64, 94, 110, 133, 178, 198, and 308 kev. These values are in excellent agreement with those reported by Cork *et al.*[1] and by Martin *et al.*[2]

The spectrum has a small bump at 250 kev. A possible reason for this is the addition of peaks when two gamma rays or x-rays are absorbed simultaneously in the same crystal. The 198-kev gamma ray and part of the K x-rays are in coincidence and they will therefore give a small peak at about 250 kev. The size of the peak depends on the solid angle subtended by the crystal at the source. In the coincidence measurements, where it is necessary to use large solid angles, this effect sometimes is of importance. For the spectrum shown in Fig. 1, however, it is very small. The bump at 250 kev must therefore be attributed to one or several gamma-rays. When the spectrum is analyzed more in detail it is found that the 250-kev peak is broader than can be accounted for by a single gamma ray. It must therefore be concluded that the spectrum contains two or more weak gamma rays in the range 240–260 kev.

[1] Cork, Keller, Rutledge, and Stoddard, Phys. Rev. **78**, 95 (1950).
[2] Martin, Jensen, Hughes, and Nichols, Phys. Rev. **82**, 579 (1951).
[3] A. Bohr, Kgl. Danske Videnskab. Selskab, Mat.-fys. Medd. **26**, No. 14 (1952).
[4] A. Bohr and B. R. Mottelson, Kgl. Danske Videnskab. Selskab, Mat.-fys. Medd. **27**, No. 16 (1953).
[5] H. Schüler and T. Schmidt, Naturwiss. **22**, 838 (1934).
[6] K. H. Lindenberger and A. Steudel, Naturwiss. **42**, 41 (1955).
[7] Sven A. E. Johansson and S. Almquist, Arkiv Fysik **5**, 427 (1953).
[8] Sven A. E. Johansson, Iowa State College Report, ISC-431 (unpublished).

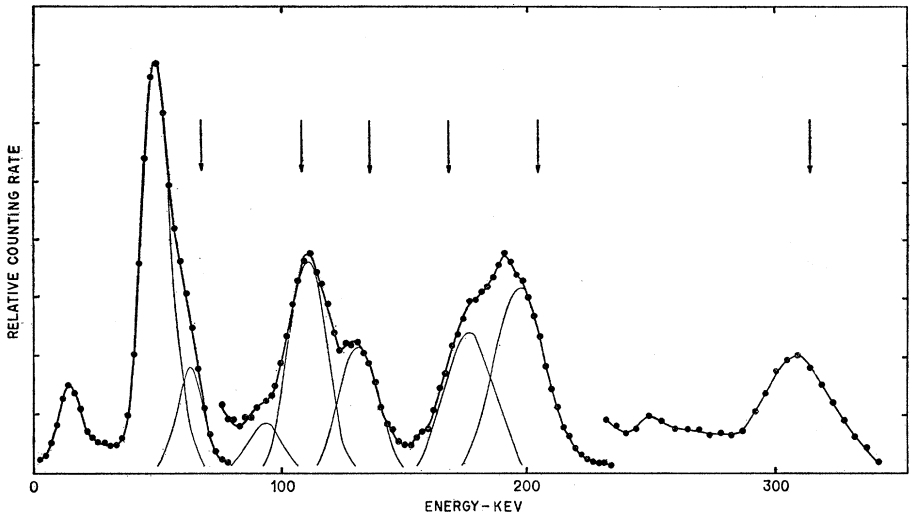

FIG. 1. Gamma-ray spectrum of Yb^{196}. The fine lines show the resolution of the spectrum into its main components.

The decay of Yb^{169} is known to proceed via a metastable state with a half-life of 0.7 μsec.[2,9] The coincidence spectrometer makes it possible to record spectra of the radiation preceding and following the metastable state. First a delay of 0.4 μsec was introduced after the spectrometer which is connected to the single-channel analyzer. The channel was set at the x-ray peak. The coincidence spectrometer will then record the spectrum of the radiation following the metastable state. It is shown in Fig. 2(a). The greater part of the radiation apparently follows the metastable state. The 64-kev gamma ray is missing, however. The broad distribution around 250 kev can be accounted for by accidental addition of the 198-kev and K x-ray peaks. Hence the weak gamma rays at 250 kev are presumably missing too.

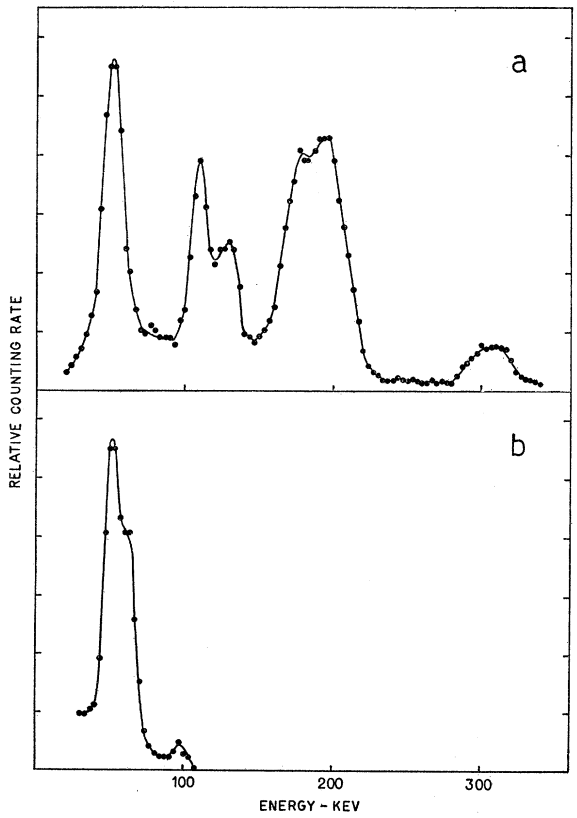

FIG. 2. (a) Spectrum of the radiation following the metastable state. (b) Spectrum of the radiation preceding the metastable state.

Next a 0.4-μsec delay was introduced after the other spectrometer in order to study the radiation preceding the metastable state. The channel of the analyzer was set on the 110-kev peak. The resulting coincidence spectrum is shown in Fig. 2(b). The only gamma rays preceding the metastable state are the ones at 64 and 94 kev.

In order to investigate the coincidence relationships a series of coincidence spectra were recorded with the analyzer channel set to select various gamma rays. Even if most of the gamma rays are not completely resolved, it is in most cases possible to find a channel position where a certain gamma ray can be practically completely separated from the adjacent rays. The various channel settings used in the present investigation are shown with arrows in Fig. 1.

First the channel was set on the 64-kev peak. The corresponding coincidence spectrum is shown in Fig. 3(a). Besides the K x-rays this spectrum contains the 94, 110, 133, 178, 198, and 308-kev gamma rays. The relative intensities are about the same as in the total spectrum with one exception: the 94-kev peak is much stronger. The only way to account for this fact is to assume that the 64-kev and 94-kev gamma rays are in prompt coincidence but that the 64-kev ray is separated from the other rays by the metastable state.

[9] S. DeBenedetti and F. K. McGowan, Phys. Rev. **74**, 728 (1948).

Since the resolving time of the coincidence circuit is shorter than the lifetime of the metastable state, the coincidence rate will be lower in the latter case. The bump at 250 kev is due to accidental coincidences in the same crystal between the 198- and 178-kev gamma rays and the K x-rays.

The channel was then set on the 110-kev peak. The coincidence spectrum is shown in Fig. 3(b). The 64-, 94-, and 178-kev gamma rays are relatively weaker than the 198-kev ray. The 110- and 133-kev rays are very weak; their presence is very likely due to secondary effects like accidental coincidences and backscattering. The spectrum also shows a bump at 260 kev. It cannot be explained as an effect of accidental coincidences as in the previous spectrum. It must therefore be assumed that the 110-kev gamma-ray is in coincidence with a weak gamma ray at about 260 kev.

Next the channel was set on the 133-kev peak. The coincidence spectrum, shown in Fig. 3(c), is similar to the previous one except that the 178-kev peak is the strongest. It is asymmetric because of the presence of a small peak at about 195 kev. If the channel setting had allowed a certain part of the 110-kev peak to fall into the channel, then the 198-kev gamma ray would have appeared in the spectrum. However, with the channel setting used, only a small part of the 195-kev peak can be due to the 198-kev gamma ray. Furthermore, if the peak is analyzed more carefully, it turns out that the small peak definitely lies lower than 198 kev, at about 194 kev. It is therefore tentatively assumed that the 133-kev gamma ray is in coincidence with a gamma ray of 194 kev. At higher energies there is a weak bump at 240 kev. As in the previous spectrum it cannot be explained as an effect of accidental coincidences. It is therefore assumed that there also exists a 240-kev gamma ray.

With the channel set on the 178-kev peak, the spectrum of Fig. 4(a) was recorded. The main gamma-ray peak is due to the 133-kev gamma ray. The 64-, 94-, and 110-kev gamma rays are relatively weaker.

Figure 4(b) shows the spectrum recorded with the channel set on the 198-kev peak. Peaks due to the 64- and 110-kev gamma rays can be seen. The 110-kev peak has a tail on the high-energy side, which must be due to a gamma ray with an energy slightly higher than 110 kev. A detailed analysis of the peak shows that the tail can be only partly ascribed to the 133-kev gamma ray. There is also a component of lower energy, which may be the 120-kev gamma ray reported by Martin *et al.*

Finally the channel was set on the 308-kev peak. The spectrum, shown in Fig. 4(c), contains the 64- and 94-kev gamma rays.

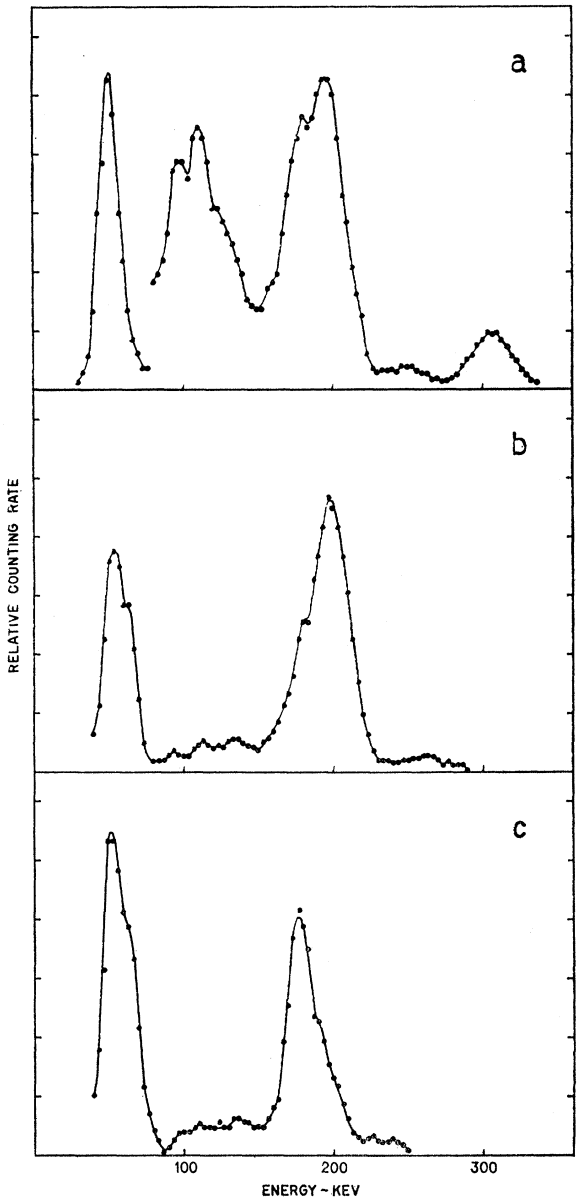

FIG. 3. Coincidence spectra with the channel set to select (a) the 64-kev gamma ray, (b) the 110-kev gamma ray, and (c) the 133-kev gamma ray.

LEVEL SCHEME

The coincidence experiments establish the level scheme shown in Fig. 5. All spectra are in agreement with this scheme. A few points warrant some discussion.

A level has been placed at 10 kev for the following reasons. The coincidence spectrum in Fig. 4(b) shows coincidences between the 198- and 120-kev gamma rays. The spectrum of the radiation preceding the metastable state at 318 kev does not contain a 120-kev gamma ray. Hence this gamma ray must follow the 198-kev gamma ray. This requires a level at 10 kev. Additional support for this interpretation is provided by the Coulomb excitation of thulium.[10] In that experiment a single

[10] T. Huss (private communication).

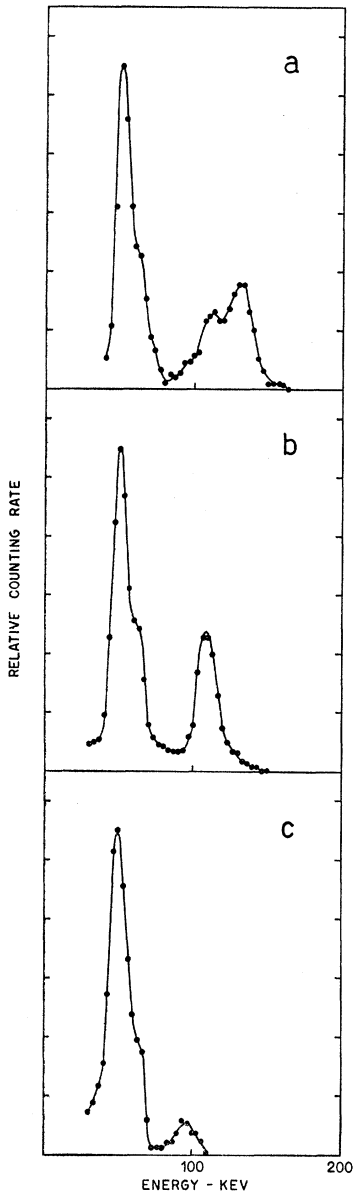

FIG. 4. Coincidence spectra with the channel set to select (a) the 178-kev gamma ray, (b) the 198-kev gamma ray, and (c) the 308-kev gamma ray.

level is excited which decays by emission of two gamma rays of 110- and 120-kev. Hence, even though the 10-kev gamma ray leading to the ground state has not been detected, there seems to be enough experimental evidence for postulating a level at 10 kev.

A gamma ray is expected to go from the 142-kev to the 120-kev level. Due to its low energy it has not been detected in the present experiment, but it is very likely the 22.8-kev gamma ray reported by Martin *et al.* This gamma ray explains some features in the coincidence experiments. Figure 3(b) shows that the 178-kev gamma ray to some extent is in coincidence with the 110-kev gamma ray. The level scheme shows that this can be explained as a cascade via the 23-kev transition. The spectrum in Fig. 4(a) gives another example of the existence of this cascade. These spectra allow a calculation of the intensity of the 23-kev transition, as will be discussed below.

The order of the 64-kev and 94-kev transitions can be determined from their relative intensities. The intensity of the corresponding gamma rays are listed in Table I. The intensity of the conversion electrons can not be determined accurately from the conversion spectrum of Martin *et al.*, but it can be seen that the 94-kev gamma ray is not especially highly converted. Hence the 94-kev transition must be considerably weaker than the 64-kev transition. The former is therefore the preceding one.

The coincidence spectra in Figs. 3(b) and 3(c) indicate that there exist two weak gamma rays at 240 kev and 260 kev in coincidence with the 133-kev and 110-kev gamma rays, respectively. These two gamma rays fit very well into the level scheme, both starting from the 382-kev level. This agreement supports the correctness of the proposed interpretation of the coincidence spectra.

All gamma rays reported are included in the level scheme, except the 142-kev and 160-kev rays. It is interesting to note that they fit well into the scheme as transitions from the 476-kev level to the 336-kev and 318-kev levels, respectively. They are so weak, however, that the coincidence measurements give no information and therefore they are not included in the scheme.

It is interesting to note that the relative intensities of the gamma rays are the same in the spectrum of the total radiation (Fig. 1) as in the spectrum of the delayed radiation. This means that no electron capture branches go to the 120-kev or 142-kev levels. It follows from the subsequent discussion that no branches go to the ground state or the first excited state.

The spectrum in Fig. 1 is used to determine the relative intensities of the gamma rays. The photoelectric efficiency of the scintillation spectrometer is calculated for the various energies, taking into account the photoelectric absorption coefficient, secondary absorption processes, K x-ray and electron escape, and absorption of the gamma rays in the material surrounding the crystal. The efficiency varies fairly slowly from about 95% at 100 kev to about 40% at 300 kev. The area of the peak times the efficiency then gives the relative intensities.

The relative intensity of the K-conversion lines has been determined by Martin *et al.* The absolute intensity, i.e., the intensity relative to the number of disintegrations, can be determined in the following way: The K x-rays are produced in two different processes, namely in the K-capture decay and in the internal conversion of the gamma rays. Hence the total intensity of the x-rays is equal to the intensity of the x-rays following the K-capture plus the intensity of the internal conversion x-rays. The total intensity of the x-rays is known relative to the gamma-ray intensities from Fig. 1. The number of disintegrations is assumed

to be equal to the sum of the intensities of the transitions to the ground state and the first excited state. This assumption follows from the fact that no K-capture branches are going to these states. The only unknown quantity in the equation is then the ratio between the relative scales for the intensities of the gamma rays and internal conversion lines. When this ratio is determined by solving the equation, all intensity values can be expressed on the same relative scale. The absolute intensities and K-conversion coefficients can then be easily obtained.

This calculation is necessarily fairly approximate. Errors in the relative intensities will cause a considerable uncertainty in the absolute values. The intensities of the K-conversion electrons from the 94-kev and 64-kev gamma rays are not known and they are therefore not included in the calculation. The calculated electron intensities and conversion coefficients are for this reason somewhat too high. This calculation, however, is mainly of interest for determining the multipolarity of the gamma rays. The accuracy is certainly enough for this purpose.

Table I shows the intensities and conversion coefficients of the main gamma rays. The multipolarities are listed in column five and the corresponding theoretical conversion coefficients[11] in column six. A comparison shows that the multipole assignments can be made without any ambiguity.

The intensity of the 23-kev transition can be calculated from the coincidence spectra in Figs. 3(b) and 4(a) as discussed above. It turns out to be 0.20. It is then possible to check the intensity values by comparing the sum of the intensities of the transitions going to a certain level with the sum of the intensities of the transitions starting from that level. Satisfactory agreement is obtained for the 120-kev and 142-kev levels, the only ones where this check is possible.

The experimental data make it possible to assign spin values to most of the levels. The only values which give agreement with the experiments are the ones shown in Fig. 5.

TABLE I. Intensity values relative to the disintegration rate, approximate K-conversion coefficients, multipole assignments, and the corresponding theoretical conversion coefficients.

Energy kev	Gamma-ray intensity	K-electron intensity	K-conversion coefficient	Multipole assignment	Theoretical conversion coefficient
K x-rays	1.13				
64	0.25				
94	0.034				
110	0.144	0.47	3	$M1$	2.4
133	0.091	0.05	0.5	$E2$	0.48
178	0.146	0.12	0.8	$M1$	0.60
198	0.214	0.09	0.4	$M1$	0.45
308	0.060	0.004	0.06	$E2$	0.05

[11] Rose, Goertzel, Spinrad, Harr, and Strong, Phys. Rev. 83, 79 (1951).

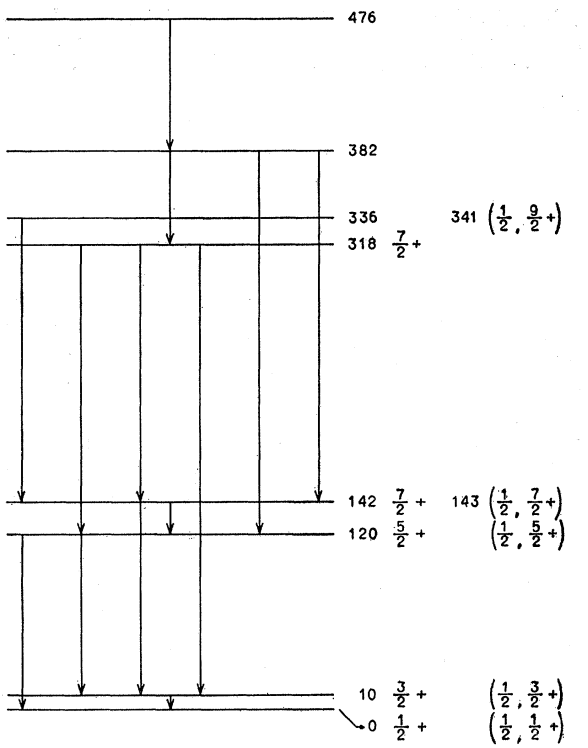

FIG. 5. Proposed level scheme of Tm^{169}. The energy and spin values to the left are experimental values. The energy values to the right are calculated from the theoretical formula with the energy values of the first two excited states inserted to determine the two parameters of the formula. The numbers in parentheses are theoretical values of K and I.

The ground state and the four lowest excited states have the same parity. The experimental material gives no information about its sign. However, both the shell model and the calculations on the wave functions in a deformed nucleus[12] indicate that the ground state is an $s_{\frac{1}{2}}$ state. Hence it is assumed that all the levels in question have even parity.

DISCUSSION

The nucleus Tm^{169} lies halfway between two closed shells and is expected to be strongly deformed. According to the unified model, the intrinsic motion of the nucleus can be separated from the collective rotational motion. Each intrinsic state is then the first member of a rotational band having states of spin

$$I=K,\ K+1,\ K+2,\ \cdots,$$

where K is the component of the total angular momentum along the nuclear symmetry axis. In the case of Tm^{169}, $K=\frac{1}{2}$ and there will be a rotational band with

$$I=1/2,\ 3/2,\ 5/2,\ \cdots.$$

[12] S. G. Nilsson, Kgl. Danske Videnskab. Selskab, Mat.-fys. Medd. (to be published).

When $K=\frac{1}{2}$ there may occur a partial decoupling of the intrinsic spin of the last odd nucleon from the rotational motion. This gives an extra rotational energy, which implies an anomalous spectrum:

$$E_I=\frac{\hbar^2}{2\Im}\{I(I+1)+a(-1)^{I+\frac{1}{2}}(I+\tfrac{1}{2})\},$$

where $\Im$ is the moment of inertia and a the decoupling parameter.

A comparison with the level scheme shows that the four first levels have the spin values expected for the rotational band. The energy formula contains two parameters which are determined by inserting the energy values of the first two excited states in the formula. The energy values of the higher levels are then obtained directly from the formula. They are shown in Fig. 5. A small correction term with negative sign has to be applied to account for the variation of the moment of inertia with I. It is proportional to $I^2(I+1)^2$ and is of importance only for the higher members of a rotational band. For the 9/2 level the correction term can be estimated to be 1–2 kev. A comparison between experimental and theoretical energy values shows a satisfactory agreement. It should be remarked that a small error in the energy values of the first two excited states can account for the small difference between experiment and theory. A better agreement is obtained by adjusting the formula to all levels by the least-squares method.

Hence the properties of the rotational band are correctly predicted by the unified model. The rotational energy quantum $\hbar^2/2\Im$ has the value 12.66 kev. Approximately the same value has been found for rotational bands in neighboring nuclei. The decoupling parameter a is equal to -0.74. A theoretical value has been calculated by Mottelson and Nilsson[13] based on wave functions obtained by considering particle motion in an axially symmetric deformed potential with the inclusion of an appropriate spin-orbit force.[12] The theoretical value is -0.8, a very satisfactory agreement.

The levels at 318, 383, and 476 kev apparently do not belong to the rotational band. The radiation probability of these levels shows a peculiar behavior. The 318-kev level decays by $M1$ and $E2$ emission but the lifetime is as long as 0.7 μsec. The 382-kev level decays mainly to the 318-kev level. One would expect that the transition probability for a transition to the 142-kev level would be considerably greater than to the 318-kev level. Similarily the 476-kev level decays to the 382-kev level but not to the lower levels. This can be explained as cases of K-forbiddenness, which means that the change in K is greater than the multipolarity of the transition.[14] In the decay of the 318-kev level, K is changed by 3 units. The lifetime according to the single-particle formula[15] is about 5×10^{-12}. Hence the retardation due to K-forbiddenness is 10^5. In the decay of the 382-kev level, one expects the intensity ratio between the 240-kev and 64-kev gamma transitions to be 100 or higher, depending on the multipolarity of the two gamma rays. Instead the 64-kev gamma ray is about 50 times as intense as the 240-kev gamma ray. The forbiddenness factor is in this case of the order 10^4 or higher.

Another possible example of K-forbiddenness is provided by the branching ratios of the K-capture. There is no branching to the 142-kev level or lower levels. The great change in K implied in transitions to those levels may account for this retardation.

The reduced transition probability for $M1$ radiation within a rotational band with $K=\frac{1}{2}$ depends on the intrinsic structure of the nucleus. When the wave functions are known the transition probabilities can be calculated and compared with the experimental values. Mottelson and Nilsson[13] have made such a calculation for the intensity ratio of the 133-kev and 23-kev gamma rays, which is found to be 20. The total intensity of the 23-kev transition has been determined as described above. With the use of the theoretical L-conversion coefficients of Rose and Goertzel,[16] one can calculate the intensity of the gamma-radiation. The intensity ratio between the 133-kev and 23-kev gamma rays then turns out to be 14, in very satisfactory agreement with the theory.

Hence the conclusion of the present work is that the level scheme of Tm^{169} makes possible a very detailed test of the unified model. There is very good agreement between theory and experiment.

ACKNOWLEDGMENTS

The author is greatly indebted to Dr. A. Bohr, Dr. B. R. Mottelson, and Dr. S. G. Nilsson for interesting discussions.

[13] B. R. Mottelson and S. G. Nilsson, Z. Physik **141**, 217 (1955).

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

[16] M. E. Rose and G. H. Goertzel, *Beta- and Gamma-ray Spectroscopy*, edited by K. Siegbahn (North-Holland Publishing Company, Amsterdam, 1955), Chap. 14.