Odd-Parity Rotational Bands in Even-Even Nuclei

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The low-intensity radiations accompanying the alpha decay of Th^{230} and Th^{228} have been studied with a scintillation and coincidence spectrometer. In addition, the alpha-particle spectrum of Th^{228} has been reexamined using a magnetic spectrograph. New gamma rays of 253 kev (8×10^{-4}%), 110 kev (1×10^{-4}%), 206 kev (5×10^{-6}%), and 235 (5×10^{-6}%) have been found associated with Th^{230} decay. A new gamma ray of 205 kev (0.03%) has been found in Th^{228} decay, and also a fifth alpha group populating a level 289 kev above the ground state was observed in an abundance of 0.03%. A new level at 416 kev in Ra^{226} suggested by these data and the coincidence measurements has been interpreted as the 6+ member of the ground-state rotational band. The remaining new levels have been assigned as 3− and 5− members of rotational bands based on the 1− states previously observed in both Ra^{226} and Ra^{224}.

INTRODUCTION

WE have found[1–3] that in a number of even-even nuclei in the radium-thorium region there exist low-lying levels with spin 1 and odd parity (1−). The excited states of two of these species, Ra^{224} and Ra^{226}, have now been studied more intensively and additional levels have been revealed which are best assigned to members of rotational bands based upon the 1− states and to higher members of the well-known bands based upon the 0+ ground states. The spectra were excited through the alpha decay of Th^{228} and Th^{230}, respectively.

EXPERIMENTAL

Th^{230} Decay

The proposed decay scheme for Th^{230} is shown in Fig. 1. The levels of Ra^{226} up to and including that at 253 kev were known previously and information on internal conversion and angular correlations has led to unambiguous spin and parity assignments.[2–8] In brief, the 68-kev and 142-kev transitions have been shown to be $E2$ transitions in cascade and define the 2+ and 4+ states relative to the 0+ ground state; the 184-kev and 253-kev transitions were shown to be parallel $E1$ transitions originating from the 1− state.

The first indication of levels above the 253-kev state came from the appearance of L x-ray coincidences with a gamma ray of 253 kev. On the assumption that the L x-rays arise from a highly converted γ ray, it could be inferred that five percent of the intensity of the 253-kev γ ray is in cascade with this transition. Also in coincidence with γ_{253} was a γ ray of 68 kev, presumably the unconverted portion of the transition giving rise to L x-rays. By critical absorption measurements the energy was bracketted between the K edges of tantalum and tungsten, 67.5–69.5 kev. These measurements demand the presence of a state at 320 kev, but could not reveal the predominant order of de-excitation: 320→253→0 or 320→68→0. Further experiments have given evidence that at least most of the 68-kev transition in coincidence with the 253-kev γ ray represents the 2+ →0+ transition; hence the coincident γ_{253} leads from the new 320-kev state to the 2+ state.

In principle, the sequence of the 253-kev and 68-kev transitions could be deduced by focusing attention on the 184-kev gamma ray and measuring whether or not it is in coincidence with slightly more than one 68-kev transition, or simply whether there is ever more than one 68-kev gamma ray in coincidence. Because of extremely low intensities it was not possible to make such measurements. However, similar information could be obtained indirectly from the L x-rays resulting from internal conversion. If the 253-kev level is not populated appreciably by a transition from the 320-kev state, then the only L x-rays in coincidence with γ_{184} would be those from the 2+ →0+ transition. As an internal intensity standard, the 142-kev transition was adopted, since it is likely that the 210-kev (4+) state is not appreciably populated from higher levels. The intensity

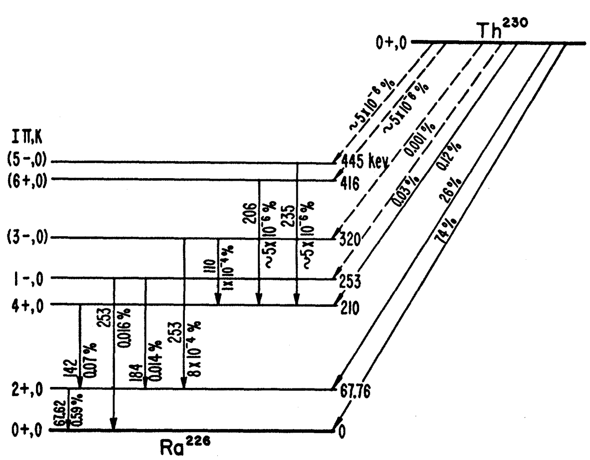

FIG. 1. Decay scheme of Th^{230}.

[1] Asaro, Stephens, and Perlman, Phys. Rev. **92**, 1495 (1953).
[2] Stephens, Asaro, and Perlman, Phys. Rev. **96**, 1568 (1954).
[3] Stephens, Asaro, and Perlman, Phys. Rev. **100**, 1543 (1955).
[4] F. Rasetti and E. C. Booth, Phys. Rev. **91**, 315 (1953).
[5] G. M. Temmer and J. M. Wyckoff, Phys. Rev. **92**, 913 (1953).
[6] Valladas, Teillac, Falk-Vairant, and Benoist, J. phys. radium **16**, 125 (1955).
[7] P. Falk-Vairant and G. Petit, Compt. rend. **240**, 296 (1955).
[8] Booth, Madansky, and Rasetti, Phys. Rev. **102**, 800 (1956).

of γ_{184} was measured relative to γ_{142}, and the L x-ray coincidence rate for each gamma ray was determined. In this way it was determined that within a standard deviation in measurement of two percent there are no L x-rays in coincidence with γ_{184} beyond those there should be from its population of the 2+ state. If all of the L x-rays in coincidence with γ_{253} came from the transition from the 320-kev state to the 253-kev state there would have been a five percent greater L x-ray$-\gamma_{184}$ coincidence rate.

Recapitulating, it would appear that the evidence points to the de-excitation of the 320-kev level principally by a 253-kev gamma ray to the 2+ state. As indicated in Fig. 1, the intensity of this γ_{253} is about five percent of the γ_{253} leading from the 1− state. The 3− assignment was arrived at for reasons which will be developed below.

Additional photons in very low intensity were found in coincidence with γ_{142} (4+ →2+ transition): 110±5 kev ($1\times10^{-4}\%$), 206±5 kev ($\sim5\times10^{-6}\%$), and 235±5 kev ($\sim5\times10^{-6}\%$). The intensities refer to percentages of the total alpha-decay events of Th^{230} and are based on the value 0.12%[9] for the alpha population to the 210-kev level. It should be pointed out that in order to see gamma rays in such low intensity it was necessary to make measurements within a few days after chemical purification of the Th^{230}, even though the daughters grow in with a 1622-year half-life. Also it might be mentioned that as a result of Compton scattering of the 253-kev gamma ray an apparent 110-kev photon is found in coincidence with one at 142 kev if the two crystals are allowed to see each other. The true gamma ray of 110 kev was observed after guarding against this effect. Owing to the low intensity of these gamma rays there were statistical problems in addition to the difficulties mentioned above, so that the energies and intensities of these gamma rays are not accurately known. Their existence, however, seems reasonably well established.

The 110-kev gamma ray agrees well with the spacing between the levels of 320 kev and 210 kev. The gamma rays of 206 and 235 kev in coincidence with the 142-kev gamma ray were used to define levels at 416 and 445 kev (Fig. 1), although from these experiments alone these assignments are by no means unique. The levels and their spin and parity designations are tentatively assigned as indicated in Fig. 1 by their agreement with expected Bohr-Mottelson rotational levels.[10,11]

If we consider the levels at 253, 320, and 445 kev to be members of a rotational band, the spins are calculated to be 1, 3, and 5, respectively, from the rotational expression:

$$E=W_0+(\hbar^2/2\mathscr{I})I(I+1).$$

This agrees, of course, with the fact that the 253-kev level is known to be 1−, but in addition the observed transitions from the other levels are consistent with their 3−, 5− designations. The 320-kev level (3−) decays to the 2+ and 4+ states as would be expected, and the 445-kev level (5−) decays to the 4+ state. No decay to the 0+ (ground) state could be detected from either of these levels, and the limit that could be set on any radiation between 300 and 700 kev was less than $7\times10^{-6}\%$.

Additional evidence in favor of these assignments may be derived from the reduced transition probabilities for the 110- and 253-kev gamma rays arising from the 320-kev level. If, in terms of the Bohr-Mottelson model, we introduce the quantum number, K, which represents the projection of the spin on the nuclear symmetry axis, then for a state of spin 1, K values of 1 and 0 are possible. In a previous publication,[3] we have used reduced transition probabilities for gamma-ray emission from the 1− states to the 0+ and 2+ states to show that in every case, a K value of 0 is clearly indicated. Thus, if the 3− state is a member of the rotational band based on this 1− state, we would expect K to be 0 for this level as well. Under these conditions the ratio of reduced $E1$ transition probabilities, γ_2/γ_4 (where γ_x represents the transition to the state of spin x), should be 0.75. The experimental ratio of 0.7 is in excellent agreement with this value. Considering the low limit on transitions to the ground (0+) state, the only other spin assignments possible are: $I=3$, $K=1$, $\gamma_2/\gamma_4=1.33$; $I=4$, $K=0$, $\gamma_2/\gamma_4=1.10$; and $I=4$, $K=2$, $\gamma_2/\gamma_4=0.34$. None of these values are in as good agreement with the data as the $I=3$, $K=0$ assignment.

The 5− state (445 kev) would not be expected to decay appreciably to the 6+ state (416 kev; see the following paragraph) due to the small energy difference (29 kev). Decay of this level to the 3− state (320 kev) would be by a rather highly converted $E2$ transition and would be difficult to detect. If the rotational transition to the 3− state does occur in appreciable intensity, the alpha population indicated to the 5− level in Fig. 1 would be too low.

The 416-kev level is very likely the 6+ member of the rotational band based upon the ground state. In this region the energy-level spacing of this rotational band is wider than in still heavier isotopes, and also the deviations from the simple $I(I+1)$ dependence given above are considerably larger. For example, using the 67.76-kev spacing of the 2+ state and the $I(I+1)$ dependence, the 4+ state would be expected at 226 kev, 16 kev higher than is found. Similarly, if we add a second term to the equation so that it becomes $E=AI(I+1)-BI^2(I+1)^2$, where $A=\hbar^2/2\mathscr{I}$, we can use the 2+ and 4+ energies to fix the constants and would then calculate the 6+ state to be at 388±10 kev, considerably lower than is found. In order to fit the three energy levels, it is necessary to add a third term, $+CI^3(I+1)^3$, to the above equation, in which case the

[9] G. Valladas and R. Bernas, Compt. rend. **236**, 2230 (1953).
[10] A. Bohr and B. R. Mottelson, Phys. Rev. **89**, 316 (1953); **90**, 717 (1953). A. Bohr and B. R. Mottelson, Kgl. Danske Videnskab. Selskab, Mat.-fys. Medd. **27**, 16 (1953).
[11] A. Bohr, *Rotational States of Atomic Nuclei* (Ejnar Munksgaard, Copenhagen, 1954).

constants may be evaluated to be

$$A = 11.74 \pm 0.10, \quad B = 0.080 \pm 0.010,$$
$$C = 0.00085 \pm 0.00025.$$

Th^{228} Decay

As seen from Fig. 1 and Fig. 2, the first three excited states of Ra^{224} are much like those of Ra^{226}; only the level spacing of the 0+, 2+, and 4+ sequence is greater and the 1− state is lower, bringing the 4+ state above the 1− state. It so happens that the 4+ state in Ra^{224} is at 253 kev, which is the same energy above the ground state as the 1− state is in Ra^{226}. The spin and parity assignments of these levels and the alpha spectrum populating the states have been discussed in earlier publications.[1,2] The characteristics of the gamma-ray spectrum as then known are the cascading $E2$ transitions from the $4+ \rightarrow 2+ \rightarrow 0+$ sequence and the branched $E1$ transitions from the 1− state to the 2+ and 0+ states.

A careful examination of the alpha spectrum revealed a fifth alpha group in 0.03% abundance populating a state 289 kev above the ground state. An upper limit of 0.01% was set for the existence of alpha groups to still higher levels.

In order to detect possible new gamma rays, gamma-gamma coincidences were measured using the 84-kev gamma ray ($2+ \rightarrow 0+$ transition) as a gating pulse. As before, the 84-kev gamma ray was found to be in coincidence with the 169-kev gamma ray ($4+ \rightarrow 2+$) and the 133-kev gamma ray ($1- \rightarrow 2+$), but in addition a weaker group at 205 ± 5 kev was seen. The sum of the 205-kev and 84-kev gamma transitions agrees well with the energy of the new state at 289 kev defined by the alpha spectrum. The abundance of the 205-kev gamma ray is the same as the alpha population of the 289-kev state within experimental uncertainty, which means that the 205-kev transition is not heavily converted and is therefore $E1$ or $E2$.

The best interpretation of the 289-kev level is that it is a 3− state. Its spacing above the 1− state is 72 kev as compared with 67 kev for the corresponding states in Ra^{226}. The 3− state decays to the 2+ state, while the branching to the 4+ state is unobservable because of the small energy difference (36 kev). It should be mentioned that a 2− assignment is ruled out because this state is populated directly in alpha decay and there can be a change in parity only for odd-integral changes in spin.

A careful search was made in the "singles" spectrum for gamma rays greater than 217 kev ($1- \rightarrow 0+$ transition). None was found and in particular it can be stated that any gamma ray greater than 275 kev must be in less than 0.001% abundance. This may be taken as additional evidence for the 3− assignment of the 289-kev state, although by itself the argument is weak.

In searching for higher levels, an additional experiment was carried out in which the 169-kev peak was

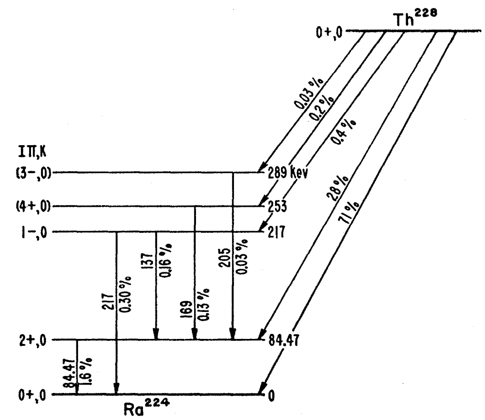

FIG. 2. Decay scheme of Th^{228}.

used to trigger the coincidence circuit. In this case the only peak observed was a very questionable one of low intensity ($7 \times 10^{-5}\%$) at 234 ± 10 kev. (The 84-kev photon would not have been seen in this experiment.) It should be added that the relatively short half-lives of Th^{228} daughters limited the time over which rare transitions could be observed. Purified samples were measured within 5 to 10 minutes and interfering daughter activities were present within one hour.

The transitions expected to be in coincidence with the 169-kev gamma ray are those from 5− and 6+ states, since these would be expected to decay to the 4+ state. The 234-kev gamma ray, if real, probably represents the $6+ \rightarrow 4+$ transition, because the energy of the level (487 kev) lies approximately where the 6+ state is expected. It is interesting to note that this state is an order of magnitude more heavily populated than the corresponding state in Th^{230} decay, and the other excited states from Th^{228} decay are also more heavily populated. The explanation for such differences is among the unsolved features of the alpha-decay process. The 5− state of Ra^{224} has not yet been observed; however, the limit of detection was not sufficiently low to make this point troublesome.

DISCUSSION

There has not yet been advanced a verifiable explanation for the appearance of low-lying 1− states in even-even nuclei. Because many of these levels lie at energies of only 200–300 kev, and because they seem to occur systematically over a region of at least twenty mass numbers, it seems likely that these states have collective rather than single-particle nature. It has been suggested[12] that the spin and parity requirements could be satisfied if the spheroidal nucleus could undergo distortions such that there be no reflection plane of symmetry perpendicular to the symmetry axis; that is, a nucleus which could assume a pear or egg shape. The

[12] Bohr, Froman, and Mottelson, Kgl. Danske Videnskab. Selskab, Mat.-fys. Medd. **29**, No. 10, 8 (1955).

fact that K, the projection of the spin on the symmetry axis, is zero for the $1-$ states (and also apparently zero for the $3-$ state in Ra^{226}) is consistent with such a model.

In terms of the theory of nuclear rotational levels, two puzzling aspects have arisen relative to these odd-parity bands. First, the spacing of these levels in a particular nucleus is such that the apparent moment of inertia is much higher than that for the ground-state configuration. It is interesting to note, although probably accidental, that the actual values for these odd-parity bands in Ra^{224} and Ra^{226} are very nearly the same as is found for the ground state of the very heavy nuclei. The second feature requiring explanation is that the relative spacing of the odd-parity levels seems to follow more closely the simple $I(I+1)$ dependence, whereas the even-parity bands in the same nuclei show easily discernible departures.

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Cross Sections for Pickup Reactions of 14-Mev Neutrons with N^{14}†

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The absolute differential cross sections for the reactions $N^{14}(n,d)C^{13}$ and $N^{14}(n,d)C^{13*}$(3.68 Mev) were measured with 14-Mev neutrons at seven laboratory angles from 0° to 65°. The deuterons were detected with a coincidence-telescope spectrometer consisting of two proportional counters and a thin CsI(Tl) scintillator. The angular distributions are both fitted with Butler-type curves for angular momentum transfers of one in agreement with positive parity and spin one for the ground state of N^{14}. The reduced width for the ground state transition agrees with that for the reaction $N^{14}(p,d)N^{13}$ as required by charge symmetry of nuclear forces. The ratio of the reduced width for the ground state transition to that for the excited state transition rules out the possibility that the N^{14} ground state be a pure $^3D_1$ state. No yield was observed corresponding to the 3.09-Mev and 3.89-Mev states of C^{13}.

INTRODUCTION

THE absolute differential cross sections for the pickup reactions of 14-Mev neutrons with N^{14} were measured as functions of angle for different deuteron groups. Figure 1 shows an energy-level diagram for the nuclides involved in the present reactions. Energy levels (in Mev) and spin and parity assignments are taken from the review article by Ajzenberg and Lauritsen.[1] Thomas[2] has made a detailed study of these levels in C^{13} on the basis of earlier measurements. Since the spins and parities of the target and residual nucleus are believed known, the measured angular distributions can be predicted on the basis of inverse stripping theory and should serve as confirmation. Similarly, the energy releases in the pickup reactions are calculable and measurements on the deuteron groups should serve as confirmation.

Inverse stripping theory leaves two adjustable parameters to fit the observed differential cross section. One is the nuclear radius and, within limits, this may be varied to fit the form of the angular distribution. The other is the reduced width of the target nucleus for separation into the residual nucleus and a proton. After the nuclear radius is determined, the absolute value of the differential cross section determines the reduced width. However, compared with the angular distribution, the reduced width is much more sensitive to the approximations in the simple Butler theory used here.[3] The values of the reduced widths can be corrected by comparison with the results of the exact evaluations of Tobocman and Kalos.[4]

Reduced widths for low-lying levels are predicted by the various coupling schemes proposed for light nuclei. Experimental determinations of reduced widths serve, therefore, as tests for these schemes. In addition, in the present case, reduced widths may be compared with those obtained in the pickup reaction of 18-Mev protons with nitrogen.[5] On the basis of charge symmetry, reduced widths should be the same for transitions involving corresponding levels of the residual nuclei. It is of some interest to check this prediction experimentally.

EXPERIMENTAL METHOD

The source of neutrons was a thick tritium target absorbed in zirconium and bombarded with 350-kev deuterons. Neutrons given off at 90° to the deuteron beam were used. These neutrons have an energy of 14.10±0.05 Mev. The associated alpha particles, at 135° to the deuteron beam, were counted to give the absolute value of the neutron flux.

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[3] R. G. Thomas, Phys. Rev. **100**, 25 (1955).
[4] W. Tobocman and M. H. Kalos, Phys. Rev. **97**, 132 (1955).
[5] K. G. Standing, Phys. Rev. **101**, 152 (1956).