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Additional evidence for the proposed excited state at ≤ 5 eV in ^{229}Th

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Angular distributions and spectroscopic strengths from the ^{230}Th$(d,t)^{229}$Th reaction provide new independent support for the rotational band assignments which led to the proposal of an excited state at ≤ 5 eV in ^{229}Th.

From a careful study of the γ transitions accompanying the α decay of ^{233}U, Kroger and Reich[1] reported that the ground and first excited states of ^{229}Th are separated by ≤ 0.1 keV. They classified these, respectively, as the $\frac{5}{2}^+$[633] and $\frac{3}{2}^+$[631] Nilsson states. Very recently Reich and Helmer,[2] using state-of-the-art germanium detectors and more accurately known calibration energies, have reported a more stringent limit of -1 ± 4 eV for the energy difference.

A long-lived nucleus with such a closely spaced pair of levels affords a special situation where excitation may be achieved by "non-nuclear" methods; for example, optical or thermal processes. Accordingly, it is timely to report additional evidence supporting the existence and location of the $\frac{5}{2}^+$[633] and $\frac{3}{2}^+$[631] rotational bands in ^{229}Th.

In connection with investigations of nuclear octupole deformation, we have recently been studying the levels of both ^{229}Th and ^{231}Th by using (d,t) particle spectroscopy. The only known previous (d,t) study of ^{229}Th has not been published and is cited in the Nuclear Data Sheets[3] only as a private communication. It was performed with 12 MeV deuterons and spectra were recorded at only two angles. To allow reliable assignments of l-transfer values we have measured triton spectra at 17 angles between 5° and 80° with a deuteron beam energy of 17 MeV. This Rapid Communication reports our evidence for rotational bands based on the $\frac{5}{2}^+$[633] and $\frac{3}{2}^+$[631] levels in ^{229}Th. As with the γ-ray studies, our technique does not permit the resolution of the two bandheads; however, the new data provide additional strong support for two very closely spaced levels in ^{229}Th.

The ^{232}Th target was prepared by vacuum evaporation of natural thorium metal on a 20-μg/cm^2 carbon foil. The ^{230}Th target ($T_{1/2} \sim 75000$ years, isotopic purity $> 99.9\%$) was prepared by direct deposition from an isotope separator onto a thicker carbon foil and is the same target that was used in an earlier study of the ^{230}Th(d,p) reaction.[4] The thorium thicknesses, determined from elastic scattering count rates in a silicon surface-barrier monitor counter at $\theta = 30°$ during the experiments, were ~ 30 and ~ 40 μg/cm^2 for ^{232}Th and ^{230}Th, respectively.

Beams of 17 MeV deuterons from the McMaster University Tandem Accelerator were used; the reaction products were analyzed with the Enge split-pole magnetic spectrograph and detected with photographic plates. Figure 1 shows the low-energy portions of the ^{230}Th$(d,t)^{229}$Th and ^{232}Th$(d,t)^{231}$Th spectra. The resolution for the ^{231}Th spectra was typically 5.5–6.0 keV full width at half maximum (FWHM) while for the ^{229}Th spectra it was ~ 7 keV FWHM in the best cases. The larger peak widths for the ^{229}Th data are believed due to the greater thickness of carbon supporting the radioactive ^{230}Th target. The Nilsson interpretations shown in Fig. 1

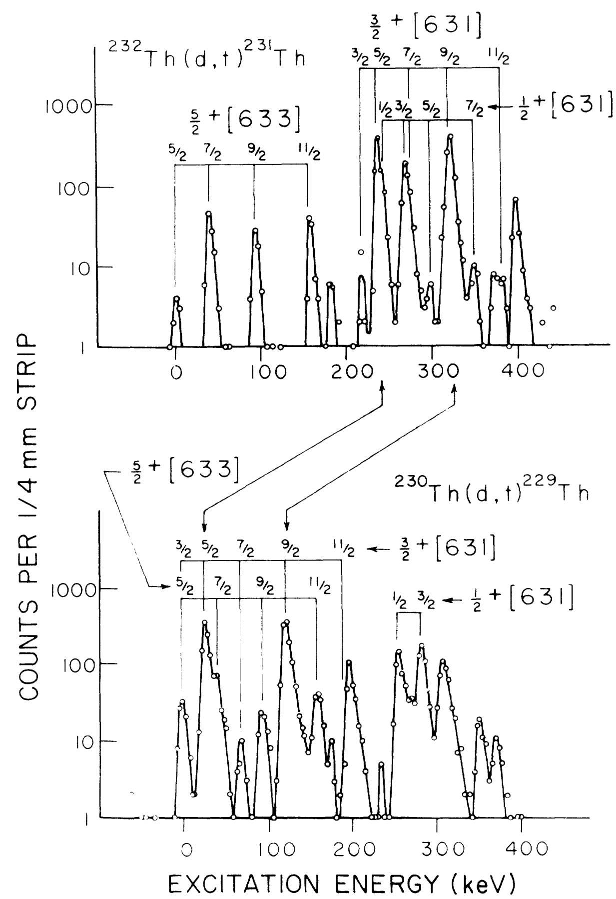

FIG. 1. Low excitation energy portions of the triton spectra at $\theta = 60°$ for the ^{230}Th(d,t) and ^{232}Th(d,t) reactions. Rotational bands are labeled with Nilsson assignments, and corresponding strongly populated $\frac{3}{2}^+$[631] band members in the two isotopes are indicated by arrows. There are many data points with zero counts between the peaks that are not shown on the logarithmic scales.

for ^{231}Th levels are taken from the extensive study of ^{231}Th by White *et al.*[4] Those shown for ^{229}Th are from the Nuclear Data Sheets.[3]

The first peak shown in the ^{230}Th$(d,t)^{229}$Th spectrum can be associated with the ^{229}Th ground-state region. This peak has a measured (d,t) Q value of -541 ± 6 keV, which agrees within error with the ground-state Q value of -537 ± 4 keV calculated from the 1986 Mass Tables.[5] Furthermore, with this first-observed peak assigned as the ground state (or the ground-state "doublet") the excitation energies for many subsequent peaks agree within ±1 keV with those of previously known levels up to several hundred keV. This excellent correspondence provides assurance that the peaks in the (d,t) spectrum have been correctly associated with the previously known levels.

Angular distributions of the (d,t) cross sections for a number of low-lying ^{229}Th levels are shown in Fig. 2, where the experimental points are compared with predicted curves from distorted-wave Born approximation (DWBA) calculations. The calculations were performed with the computer program[6] DWUCK4, using optical model parameter set CP from Wilcke *et al.*,[7] except that the recommended[6] finite range parameter (0.845) and nonlocal parameters (0.54 for deuteron and 0.25 for triton) were also included. These parameters do not significantly change the shapes of the angular distributions, or the relative cross sections for different l values, but their use removes a large part of the problem reported by Wilcke *et al.* that the observed cross sections were all larger than the calculated ones.

It can be seen from Fig. 2 that the angular distributions have quite distinctive shapes for l values of 0, 1, 2, and 4. Hence, for strong transitions with good statistics it is possible to determine l values from the angular distributions. In particular, the strongly populated levels at 29 and 125 keV were assigned by Kroger and Reich as $I^\pi=\frac{5}{2}^+$ and $\frac{9}{2}^+$ members of the $\frac{3}{2}^+[631]$ band, and the results of Fig. 2 show the (d,t) transitions to these levels have $l=2$ and 4, respectively, thus supporting those assignments. Similarly, the levels at 262, 289, and 535 keV are seen to have l values of 0, 2, and 1, respectively, consistent with the earlier interpretations.

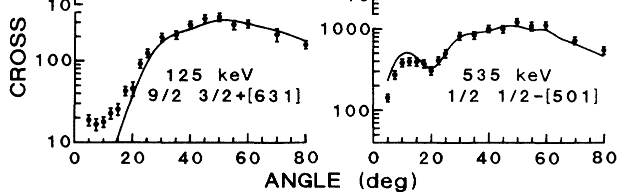

FIG. 2. Angular distributions for (d,t) cross sections of some ^{229}Th levels. The solid curves are DWBA calculations for the l values appropriate for the states indicated, adjusted in the vertical direction to give the best visual fit to the data points.

One of the most powerful tests that can be made by using single-nucleon transfer reactions, however, is to consider the relative cross sections for various spin members of a rotational band. The principles have been extensively discussed in several review articles[8-10] and will not be repeated in detail here. The (d,t) cross section on an even-even deformed target nucleus for population of a rotational band member of spin $I=j$ is

$$\frac{d\sigma}{d\Omega}=2C_{jl}^2V^2N\left(\frac{d\sigma}{d\Omega}\right)_{\rm DW}. \quad (1)$$

The C_{jl} values are wave function coefficients for the Nilsson state on which the band is based, and V^2 is the probability that this orbital is occupied by a neutron pair in the target nucleus. N is a normalization factor for the DWBA cross sections, $(d\sigma/d\Omega)_{\rm DW}$, and has been given the standard value[6] of 3.33. Since each Nilsson orbital has its particular set of C_{jl} coefficients, there is a characteristic set of cross sections to the various rotational band members. Furthermore, if there is no major change in nuclear shape between two nuclides, the orbital wave functions and hence the patterns of cross sections should be similar for a particular band in the two nuclides. In the present case, observed cross section patterns for important low-lying bands in ^{229}Th can be compared with those for the corresponding bands in ^{231}Th, where the assignments are known with some confidence.[4] This provides a rather sensitive test of the previous band assignments in ^{229}Th.

The most important question is whether the $\frac{5}{2}^+[633]$ and $\frac{3}{2}^+[631]$ bands assigned by Kroger and Reich with bandheads at ~0 keV are supported by the observed patterns of cross sections. From Fig. 1 it is seen that in ^{231}Th there are no members of the $\frac{5}{2}^+[633]$ ground-state band that are populated with large cross sections. On the other hand, the $\frac{3}{2}^+[631]$ band has rather large peaks for the spin $\frac{5}{2}^+$ and $\frac{9}{2}^+$ members, with all others being weak. For ^{229}Th, it is seen immediately from Fig. 1 that the dominant peaks in the low-energy region of the spectrum are for the levels at 29 and 125 keV, previously assigned as the $\frac{5}{2}$ and $\frac{9}{2}$ members of the $\frac{3}{2}^+[631]$ band. This is therefore very similar to the situation described above for ^{231}Th, as expected if the earlier assignments are correct. In order to make a quantitative comparison of the (d,t) strengths in the two nuclides, experimental values of the ratio $(d\sigma/d\Omega)/[2N(d\sigma/d\Omega)_{\rm DW}]$ for the various band members are listed in Table I. From Eq. (1) it is seen that this ratio should be equal to $C_{jl}^2V^2$ for each j value. The values for ^{229}Th are strikingly similar to those for ^{231}Th, and are therefore good evidence that the band assignments for ^{229}Th are correct. Although it is not possible to resolve the proposed bandheads in this experiment it is noted that the small strength observed for the ground-state doublet in ^{229}Th is very similar to the sum for the two bandheads in ^{231}Th.

The final column of Table I shows theoretical Nilsson model coefficients C_{jl}^2 for these bands, calculated with deformations $\epsilon_2=0.18$, $\epsilon_4=-0.05$, and Nilsson model pa-

TABLE I. Comparison of (d,t) strengths for rotational band members in ^{229}Th and ^{231}Th.

Rotational band member	Excitation energy (keV) ^{229}Th	Excitation energy (keV) ^{231}Th	Observed strengths $C_{jl}^2 V^2$ ^{229}Th	Observed strengths $C_{jl}^2 V^2$ ^{231}Th	Theoretical C_{jl}^2
			$\frac{5}{2}^+$[633]		
$I=\frac{5}{2}$	~0	0	≤0.009[a]	0.0017	0.010
$\frac{7}{2}$	42	42	~0.06	0.062	0.062
$\frac{9}{2}$	97	96	0.024	0.038	0.21
$\frac{11}{2}$	163	162	~0.46	0.49	0.67
			$\frac{3}{2}^+$[631]		
$I=\frac{3}{2}$	~0	221	≤0.009[a]	0.0064	0.001
$\frac{5}{2}$	29	241	0.10	0.11	0.068
$\frac{7}{2}$	72	275	~0.014	≤0.07[b]	0.010
$\frac{9}{2}$	125	325	0.45	0.54	0.49
$\frac{11}{2}$	196	386	≤0.4[b]	0.08	0.38

[a]This upper limit for the strength is obtained assuming the full cross section of the unresolved doublet is for this level.
[b]Upper limit only. Peak is obscured by a larger one for a nearby level.

rameters $\kappa=0.050$ and $\mu=0.448$ as recommended by White *et al.*[4] The observed strengths are in approximate agreement with the theoretical patterns of C_{jl}^2 coefficients, when one considers that the fullness parameter V^2 would be expected to have a value slightly larger than $\sim\frac{1}{2}$ for all four bands in Table I (since these orbitals should be near or just below the Fermi surface). In view of the simplified description used here for the nuclear model and the reaction mechanism, the agreement between the measured and expected strengths is considered to be quite good. For example, the patterns of strengths are known to be somewhat dependent on the Nilsson model parameters used,[4] and on the details of the reaction process.[7] Also, the relative strengths can often be affected by Coriolis mixing, which has been ignored here because previous works[1,4,7] suggest that the perturbations are not large for the levels considered in Table I.

It must be asked whether there is any other interpretation for the proposed $\frac{3}{2}^+$[631] band members that would be consistent with the available data. Some obvious possibilities considered by Kroger and Reich[1] are that the levels at 29, 72, 125, and 196 keV could be the lowest members of a $K=\frac{5}{2}$ or a different $K=\frac{3}{2}$ band. The $l=2$, 4 transitions of this work confirm that the parity of this band is positive. The possibility of it being a $K=\frac{3}{2}$ band starting at 29 keV is made very unlikely by the present results, since the 125 keV level would then have $I^\pi=\frac{7}{2}^+$. This level has a very large (d,t) strength but there is very little $g_{7/2}$ hole strength expected in this mass region. If one considers that these levels might be a $K^\pi=\frac{5}{2}^+$ band based on the 29 keV level, the present results rule out all possibilities except that it could be a $\frac{5}{2}^+$[622] band. The C_{jl}^2 coefficients for this orbital are not too different from those of corresponding j values in the $\frac{3}{2}^+$[631] band (within the uncertainties of the Nilsson model). However, Kroger and Reich[1] have already given several arguments for rejecting such an interpretation.

The most important evidence from Table I is that the distributions of (d,t) strengths for the proposed $\frac{5}{2}^+$[633] and $\frac{3}{2}^+$[631] bands in ^{229}Th are virtually identical to those observed in ^{231}Th, where the band assignments are well established. Thus, these results give independent support for the previously assigned band structures, and hence indirectly for the proposed existence of a very low-energy (≤5 eV) nuclear excitation in ^{229}Th. These results do not provide a determination of the energy separation of the doublet, or indicate which bandhead has the lowest energy. It is hoped, however, that the additional support provided by these data will stimulate further investigations of this apparently unique ground-state nuclear doublet.

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