# K-Capture Branch in $Tm^{170+}$

PAUL P. DAY

Argonne National Laboratory, Lemont, Illinois (Received March 14, 1956)

The electromagnetic spectrum resulting from the decay of Tm<sup>170</sup> has been examined on a ten-inch bent-crystal spectrometer. One gamma ray, seven K x-rays, and one L x-ray were observed. The presence of Er K x-ray was established and shown to be due to a 0.15% K-capture branch in the decay of Tm<sup>170</sup>. The energy of the first excited state of  $Yb^{170}$  was found to be  $84.229\pm0.041$  kev. The reflectivity of the bent-topaz crystal used in the spectrometer was found to obey the function  $(1/E)^{1.35\pm0.05}$ .

#### INTRODUCTION

HE essential details of the decay of 129-day Tm<sup>170</sup> have been given by Graham, Wolfson, and Bell.<sup>1</sup> They found that it decays by the emission of two negative beta groups (968 kev and 884 kev) to the ground state and first rotational level of Yb<sup>170</sup>. The energy of this level was determined from the K, L, and M conversion electrons of the gamma-ray transition between this level and the ground state and was reported as  $84.1 \pm 0.1$  kev. They also gave upper limits for decay by K-capture of 0.3% and by positive beta-ray emission of 0.01%. The Tm<sup>170</sup> spectrum was originally taken under investigation in order to determine the reflectivity of topaz as a function of energy with greater precision than was obtained in a previous paper.<sup>2</sup>

### SAMPLE

A radiochemically and spectroscopically pure sample of Tm<sup>169</sup> which had been irradiated at a neutron flux of about 1014 neutrons/cm2/sec for 77 days was used for the ten-inch bent-crystal spectrometer source.<sup>2</sup> With a transmission of about 10<sup>-6</sup> in the fifty- to eighty-kev region, it was desirable to have a source of several millicruries in order to have counting rates of about 500 counts/minute on the line peaks in this region. An HCl solution containing about 0.1 milligram of Tm<sub>2</sub>O<sub>3</sub> was evaporated on a strip of filter paper 1 mm wide by 17 mm long. The filter paper was dried in between applications under a heat lamp. The loaded sample had a total disintegration rate of about  $3 \times 10^{10}$ per minute. The active filter paper was placed in a Lucite sample holder in such a way that the thickness of the paper (0.007 in.) served as the line-source width in the bent-crystal spectrometer. Even with so small a source width, the resolutions of 1.8% for the 84-kev gamma ray and 1.1% for the K x-rays were still limited by this width.

#### EXPERIMENTAL RESULTS

One gamma ray, seven K x-rays, and one L x-ray were observed on the bent-crystal spectrometer. Table I lists the energies and relative intensities of the lines. Columns (2) and (3) list the energies that were obtained from the first- and second-order reflections from the bent-topaz crystal, respectively. Column (5) lists the energies of the x-rays obtained from Cauchois and Hulubei<sup>3</sup> using a conversion factor of 12372.44 kev x-units given by Cohen et al.<sup>4</sup> The energy of the gamma ray listed in column (5) is that obtained by Graham et al.<sup>1</sup> Owing to the limited energy range of the experiment and the small mass of the source, it was necessary to correct the relative intensities only for reflectivity, scintillation detector escape peak, and vertical divergence.

In an experiment on the electromagnetic spectrum of Am<sup>241</sup>,<sup>2</sup> in which the reflectivity of the bent-topaz crystal was determined by a rather inaccurate method, it was found that the reflectivity more nearly obeyed a 1/E dependence rather than a  $1/E^2$  dependence as found by Lind et al.<sup>5</sup> for a bent-quartz crystal. Using the present Tm<sup>170</sup> sample and measuring the relative intensities of the gamma ray and the K x-ray peaks on a scintillation counter and then comparing the relative intensities of the diffracted peaks obtained on the bent-crystal spectrometer, a value of  $1.35 \pm 0.05$  is obtained for the reflectivity function exponent. Applying this function to the Am<sup>241</sup> data, the ratio of the sixty-kev gamma ray to the sum of all other gammas and L x-rays gave a ratio of 1.7, which agrees precisely with the scintillation detector relative intensity measurement. Although the Tm experiment measured the reflectivity only over the range from 50- to 84-key. the internal agreement obtained in the Am<sup>241</sup> experiment indicates that the 1.35 power is also applicable over the low-energy range (down to about 12 kev) of the spectrometer.

Figure 1 depicts the K x-rays of Yb and Er, indicating the resolution of the bent-crystal spectrometer and the unquestionable presence of the Er K x-rays. The ratio of the Er K x-rays to Yb K x-rays was 0.033 $\pm 0.011$ . One year after the first measurement, another sample was prepared from the original solution and a

<sup>†</sup> Work performed under the auspices of the U.S. Atomic Energy Commission.

<sup>&</sup>lt;sup>1</sup> Graham, Wolfson, and Bell, Can. J. Phys. **30**, 459 (1952). <sup>2</sup> Paul P. Day, Phys. Rev. **97**, 689 (1955).

<sup>&</sup>lt;sup>3</sup> Y. Cauchois and H. Hulubei, Constantes Sélectionnées, Long-<sup>11</sup>. Calculus and n. Infinitely, Constants Selectionness, Emg-ueurs D'Onde Des Émissions X Et Des Discontinuités D'Absorption X (Hermann & Cie, Paris, 1947).
<sup>4</sup> Cohen, DuMond, Layton, and Rollett, Revs. Modern Phys.
27, 363 (1955).
<sup>5</sup> Lind, West, and DuMond, Phys. Rev. 77, 475 (1950).

Line (1)	First order (kev) (2)	Second order (kev) (3)	Weighted average (kev) (4)	Table value (kev) (5)	Intensity (6)
Gamma	84.249±0.065	$84.216 \pm 0.052$	$84.229 \pm 0.041$	84.1±0.1	107
$Yb K\alpha_1$	$52.385 \pm 0.028$	$52.370 \pm 0.018$	$52.374 \pm 0.015$	52.363	100
Yb $K\alpha_2$	$51.358 \pm 0.032$	$51.322 \pm 0.020$	$51.332 \pm 0.017$	51.342	56
Yb $K\beta_{1,3}$	$59.308 \pm 0.035$		$59.308 \pm 0.035$	59.310	28
Yb $K\beta_{2}$	$61.021 \pm 0.065$		$61.021 \pm 0.065$	60.882	7.2
$Yb L\beta_1$	$8.39 \pm 0.1$		$8.39 \pm 0.1$	8.413	
Er Ka	$49.117 \pm 0.038$		$49.117 \pm 0.038$	49,103	3.3
Er Ka	$48.240 \pm 0.050$		$48.240 \pm 0.050$	48.209	1.9
$\text{Er } K\beta_{1,3}$	$55.515 \pm 0.125$		$55.515 \pm 0.125$	55.624	1.2

TABLE I. Electromagnetic radiation observed in the decay of Tm<sup>170</sup>.

ratio of  $0.028 \pm 0.011$  was obtained for the K x-ray ratio. In order to be sure that the two samples had approximately the same self-absorption, the L x-ray region was scanned and it was found that the relative intensity of the Yb  $L\beta_1$  x-ray in the second sample was approximately the same as that found in the first sample. Since the sample self-absorption would be much more pronounced in the L x-ray region than in the K x-ray region, it may be concluded that the two samples had essentially the same mass (probably negligible with respect to the Er to Yb K x-ray ratio). Since the Er K x-ray to Yb K x-ray ratio remained at about the same level within experimental error over a one-year period, it was concluded that the  $\operatorname{Er} K$  x-rays were associated either with the decay of Tm<sup>170</sup>, with a different Tm activity, or with a Ho negative beta activity of approximately the same half-life (129 days). Relying on the present knowledge of all the Tm and Ho isotopes, one concludes that the Er K x-rays must arise in one or more of the following ways; (1) K-capture in Tm<sup>168</sup> (the errors in the two experimental Er/Yb x-ray ratios are too large to distinguish between the 85-day Tm<sup>168</sup> and 129-day Tm<sup>170</sup> activities), (2) erbium fluorescence, (3) Ho<sup>166</sup> beta decay, or (4) K-capture in Tm<sup>170</sup>.

According to Wilkinson and Hicks,<sup>6</sup> 85-day Tm<sup>168</sup> has two gamma rays, with energies of 210 kev and 850 kev and relative intensities of 0.2 and 0.8, respectively (K x-ray intensity=1.0). In order to check on the possibility of the presence of Tm<sup>168</sup> in the sample [produced by an (n,2n) reaction], a gamma-ray pulse-height spectrum of the intense bent-crystal spectrometer sample was taken on a 20-channel scintillation spectrometer. In order to eliminate scattering of the intense 84-kev gamma ray into the detector, the source was surrounded with one inch of lead, with exception of the side facing the NaI detector which was covered with 2.65  $g/cm^2$  of lead to reduce the very intense soft radiation. The observation of the 210-kev gamma ray was rendered difficult by the presence of the bremsstrahlung radiation from the intense 884-kev and 968-kev beta rays from the Tm<sup>170</sup> decay. Over the region in which the 850-kev gamma ray should lie, there was no perceptible photopeak superimposed on the high energy tail of the bremsstrahlung radiation; the latter had an integrated counting rate of 16 counts/minute over the 850-kev region. With a source geometry of about 4%, a detector efficiency of 5%, and a half-thickness of 8.5 g/cm<sup>2</sup> of lead for 850-kev radiation, the over-all efficiency of the counting system was  $1.6 \times 10^{-3}$ . Using the Wilkinson data for the relative intensities, and assuming that the entire 16 counts/minute in the 850-kev region are due to the gamma ray, one computes that the total Tm<sup>168</sup> disintegration rate would be 13 000 counts/minute. Since the transmission of the bent-crystal spectrometer in the Er K x-ray region (49 kev) is about  $10^{-6}$ , the contribution to the observed Er x-ray peaks from the Tm<sup>168</sup> that may be present would be negligible.

According to a spectrochemical analysis of the sample, there was less than 0.02% Er present. Since the Tm K x-rays are not even observed, and Tm makes up the major portion of the sample, Er fluorescence could not be the source of the Er x-rays.

Butement<sup>7</sup> has reported a long-lived isomer of Ho<sup>166</sup>. Using this data, assuming a long half-life (such that irradiations of up to 77 days will increase the specific activity linearly), taking the spectrochemical analysis figure of 0.05% Ho, and assuming a total sample mass of 0.1 milligram, one computes that the total



FIG. 1. The K x-ray region of  $Tm^{170}$  as seen on the bent-crystal spectrometer.

<sup>7</sup> F. D. S. Butement, Proc. Phys. Soc. (London) A65, 254 (1952).

<sup>&</sup>lt;sup>6</sup> G. Wilkinson and H. G. Hicks, Phys. Rev. 75, 696 (1949).

Ho activity would amount to about 40 disintegrations/ minute. With a transmission of  $10^{-6}$  for the spectrometer, any contribution to the Er K x-rays from the Ho<sup>166</sup> activity that might be present would be negligible.

Since Graham et al.<sup>1</sup> set a 0.01% upper limit for positron emission, one is thus left with the fourth and final alternative; the Er K x-rays are due to a small K-capture branch in the Tm<sup>170</sup> decay. Taking the intensity of the Yb K x-rays as 0.094 per disintegration<sup>1</sup> and ignoring the K x-rays that might be coming from conversion on the K-capture side, the intensity of the K-capture branching ratio is calculated to be 0.15%. This value is consistent with that of Graham et al.,<sup>1</sup> who set an upper limit of 0.3%.

With the establishment of the K-capture branch, it seemed worthwhile to look for the presence of a gamma ray associated with the de-excitation of the first rotational level in Er<sup>170</sup>. From a plot of the energy of the first excited state of the even-even nuclei as a function of mass number, one would estimate that the energy of the first excited state of Er170 would lie somewhere in the region between seventy and eightyfive kev. If the K-branch proceeded entirely through the first excited state, one would expect the intensity of the gamma ray (no conversion) to be about 5%of the 84-kev gamma ray. With the present resolution of about 1.8% in this region and only a moderately intense source (84-kev gamma peak counting rate of 400 counts/min), it would be difficult to see a 5% contribution to the 84-kev gamma ray from the Er<sup>170</sup> gamma ray in the region between 82.7 and 85.7 kev. A search was made on the bent-crystal spectrometer in the 70- to 8.3-key region and the 85- to 90-kev region; there was no evidence of any gamma ray present with an intensity greater than 2% of the 84-kev gamma ray. However, systematics indicate that the gamma rays would have about the same conversion coefficients and that the branching ratios to the first excited level in each daughter nucleus would be about the same ( $\approx 20\%$ ), and thus the Er K x-rays arise mainly in the K-capture process rather than in K conversion. Thus the ratio of the two gammaray intensities should be about  $2 \times 10^{-3}$ . Therefore it is not surprising that this line was not observed.

PHYSICAL REVIEW

#### VOLUME 102, NUMBER 6

JUNE 15, 1956

## Neutron Resonances in the kev Region<sup>\*</sup>

JOHN H. GIBBONST

Department of Physics, Duke University, Durham, North Carolina (Received August 11, 1954; revised manuscript received February 13, 1956)

A neutron collimation-detection system has been constructed for use in measurements of resonance neutron cross sections. The high efficiency of the detector in combination with careful collimator design enables one to collect data with relatively good resolution in a short time. Although designed to measure cross sections at a minimum energy of about 5 kev, resonances have been observed at energies as low as 350 ev. Resolving power exceeds that of velocity selectors, however, only at neutron energies greater than about 5 kev. Resonances in iron were found at 8, 29, 75, and 85 kev. The three latter resonances exhibit strong interference dips and are assumed to be due to s-wave neutrons on  $Fe^{56}$ . The resonance energies found in bismuth were 1, 3, 13, 16, 34, 47, and 70 kev; the level spacing for a single J value thus seems to be about 20 kev.

### INTRODUCTION

HERE is now considerable evidence<sup>1-7</sup> that the effects of magic neutron numbers on nuclear level density are evident even at excitation energies of 5-8 Mev. One of the most direct methods of investi-

<sup>1</sup> Hughes, Spatz, and Goldstein, Phys. Rev. **75**, 1781 (1949). <sup>2</sup> D. J. Hughes and D. Sherman, Phys. Rev. **78**, 632 (1950). <sup>3</sup> J. H. Gibbons and H. W. Newson, Phys. Rev. **91**, 209(A) (1953)

gation of this effect is the study of neutron total cross sections in the resonance region. Many "magic" or "near-magic" nuclei have resonance spacings of the order of 1 to 10 kev. With a spectrometer of 5 to 10% resolving power in the range 5 to 100 kev one could also hope to finish bridging the gap<sup>8</sup> between neutron cross sections obtained by velocity selectors (0-5 kev) and the other principle group of data, whose low-energy side is somewhat below 100 kev. Finally, determination of resonance parameters, such as reaction width and spin of many levels previously too narrow to measure, should be feasible.

This paper reports the successful construction of a

<sup>\*</sup> This work was supported by the U. S. Atomic Energy Commission.

<sup>†</sup> Now at Oak Ridge National Laboratory, Oak Ridge, Tennessee.

<sup>&</sup>lt;sup>4</sup> H. W. Newson and R. H. Rohrer, Phys. Rev. **94**, 654 (1954); **87**, 177 (1952). <sup>5</sup> Miller, Adair, Bockelman, and Darden, Phys. Rev. **88**, 83

<sup>(1952).</sup> 

<sup>&</sup>lt;sup>8</sup> Hughes, Garth, and Levin, Phys. Rev. 91, 1423 (1953).

<sup>&</sup>lt;sup>7</sup> Harvey, Hughes, Carter, and Pilcher, Phys. Rev. 99, 10 (1955).

<sup>&</sup>lt;sup>8</sup> D. J. Hughes and J. A. Harvey, *Neutron Cross Sections*, Brookhaven National Laboratory Report BNL-325 (Super-intendent of Documents, U. S. Government Printing Office, Washington, D. C., 1952).