K Series X-Ray Wavelengths in Rare Earth Elements*

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The K-series x-rays in ten rare earth elements have been studied with a two-meter-radius bent-quartzcrystal spectrograph. The 3.7-Mev proton beam of the A-48 accelerator (UCRL, Livermore) was used to produce the atomic excitations. The wavelengths obtained for the $K_{\alpha 1}, K_{\alpha 2}, K_{\beta 3}$, and $K_{\beta 1}$ lines are compared with previous wavelength measurements. Two weaker transitions, $K_{\beta\beta}$ and $KO_{11}O_{111}$, were also observed and the energies are compared with energies obtained from tables of known atomic energy levels.

I. INTRODUCTION

HE wavelengths of nuclear gamma rays resulting from electric (or Coulomb) excitation have recently been determined with high precision using a bent-quartz-crystal spectrograph.¹ In all electric excitation experiments, the characteristic K-series x-rays of the element which is being bombarded are also observed.² Generally, these x-rays appear as a rather broad peak in the NaI crystal pulse-height spectrum which is centered around the $K_{\alpha 1}$ and $K_{\alpha 2}$ energy of the element. At a proton energy of 3.7 Mev, the total intensity of these characteristic x-rays emitted from the target may be as much as 50 to 100 times as large as the intensity of the nuclear gamma rays. (In most electric excitation experiments it is indeed necessary to use selective absorbers to reduce the x-ray intensity relative to that of the nuclear gamma rays.³)

The high resolution of the bent-quartz-crystal spectrograph makes it possible to observe directly seven lines of the K spectrum of the target element. The most intense lines are $K_{\alpha 2}$, $K_{\alpha 1}$, $K_{\beta 3}$, and $K_{\beta 1}$ which arise from the transitions between the K shell and the L_{II} , $L_{\rm III}$, $M_{\rm II}$, and $M_{\rm III}$ shells, respectively. The doublet line, $K_{\beta 2}$ is also easily visible. This line corresponds to the transitions between the K and the $N_{\rm II}$ and $N_{\rm III}$ shells. Finally, two rather weak lines also appear on each plate. One of these is the transition between the Kand the O_{II} and O_{III} shells, and the other is a line, usually called $K_{\beta 5}$, which corresponds to the quadrupole transition between the K and the M_{IV} and M_V shells. Both of these weak lines are doublets, but the resolution of the bent crystal is not sufficient to separate the components. The most intense lines $(K_{\alpha 2}, K_{\alpha 1}, K_{\beta 3}, \text{ and }$ $K_{\beta 1}$) are easily visible in an exposure time of the order of 10 ma-hr since the atomic excitation cross sections in the region Z > 60 are reasonably large.⁴ The two weak

lines, $K_{\beta 5}$ and $KO_{II}O_{III}$, have intensities between 10^{-2} and 10^{-3} of the $K_{\alpha 1}$ line, and hence exposures of the order of 100 ma-hr are necessary.5 (For a detailed discussion of the efficiency of the spectrograph and the exposure times necessary for measurements, see reference 1.)

It has already been pointed out that in the electric excitation experiments,¹ certain characteristic x-ray lines from each of the target elements were used for calibration purposes. In several cases it was found that certain lines, particularly $K_{\beta 3}$ and $K_{\beta 1}$ x-rays, in some of the rare earths seemed to give unsatisfactory results when the known⁶ wavelengths for these lines were used for calibration. These discrepancies led to a careful examination of the previous measurements of the Kemission spectra in the rare earth region, and it was therefore decided to remeasure the intense K-series x-rays in the rare earth elements. In addition, the wavelengths of the weak $K_{\beta 5}$ and $KO_{II}O_{III}$ transitions were also measured, since these have apparently never been directly observed as emission lines in the rare earth elements using the conventional method of excitation by electron bombardment in an x-ray tube. Weak emission lines in heavy elements are easy to observe, however, if protons are used for atomic excitation since the continuous Bremsstrahlung expected from high-energy protons is many orders of magnitude smaller⁷ than that produced by electrons.

II. EXPERIMENTAL PROCEDURE

The experimental methods used in this work have been described in reference 1. The weak transitions ($K_{\beta 5}$ and $KO_{II}O_{III}$) were observed on the same spectral plate which were used in reference 1 to record the nuclear gamma rays from electric excitation. The samarium Kseries x-ray spectrum is shown in Fig. 1 with both of the weak lines clearly visible on the plate. The calibration lines used to determine the wavelengths of these x-rays are shown in Table I of reference 1. The strong lines $(K_{\alpha 1}, K_{\alpha 2}, K_{\beta 3}, \text{ and } K_{\beta 1})$ were measured by recording the x-ray spectrum obtained from several different targets on the same spectral plate. Three such plates were made, each having three or four x-ray spectra

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chusetts. ¹ Chupp, Du Mond, Gordon, Jopson, and Mark, Phys. Rev. 112, 518 (1958).

^a Alder, Bohr, Huus, Mottelson, and Winther, Revs. Modern Phys. 28, 432 (1956). ³ P. H. Stelson and F. K. McGowan, Phys. Rev. 105, 1346

^{(1957).}

⁴ Lewis, Simmons, and Merzbacher, Phys. Rev. 91, 943 (1953).

⁵ P. A. Ross and F. Bloch, Phys. Rev. 47, 884 (1935).

⁶ J. M. Cork and B. R. Stephenson, Phys. Rev. **27**, 530 (1936). ⁷ S. D. Drell and K. Huang, Phys. Rev. **99**, 686 (1955).



FIG. 1. K-series x-ray spectrum of samarium. This plate was exposed for 100 ma-hr. For a part of the exposure, the lower portion of the plate was shielded with a lead screen. The weak transitions ($K_{\beta\beta}$ and $KO_{II}O_{III}$) are easily seen on the plate.

along with the calibration lines from a tantalum source, discussed in reference 1. Since exposures of only 10 ma-hr were necessary to record each spectrum, such plates could be made in reasonably short times. Figure 2 shows the plate with the x-ray spectra of dysprosium, holmium, and erbium. The strong x-ray lines were recorded in this manner so that relative distance measurements could be made in case there were large discrepancies in the absolute wavelength measurements. The methods used to calculate the wavelengths λ and standard deviations $\sigma(\lambda)$ of unknown lines are described in reference 1.

III. RESULTS

The wavelengths of all x-ray lines measured are shown in Table I. The wavelengths obtained in the present work are compared with wavelengths measured previously by several workers. The large discrepancies between the $K_{\beta 3}$ and $K_{\beta 1}$ wavelengths for certain elements given here and those given by Cork and Stephenson⁶ are not surprising. These authors state that the standard deviations of their measurements of the $K_{\beta 3}$ and $K_{\beta 1}$ lines are considerably larger than ± 0.05 x-units, and Ingelstam⁸ has estimated that their standard deviations may exceed ± 0.1 x-unit. The dysprosium x-rays were measured on two separate plates. The values obtained on the second plate are shown in parentheses in Table I. The good agreement between the two sets of numbers indicates the reliability of the method.

Table II shows a comparison between the energies of the transitions measured and the energies of the corresponding transitions obtained from existing tables of atomic energy levels.⁹ These latter tables were obtained by combining the best available measurements of the Kand L_{III} absorption edges with the best measurements of L-emission spectra.¹⁰ The comparison made here is between the levels obtained in this manner and our direct determinations of the transition energies by measurement of the corresponding wavelengths. The energies shown in Table II,¹¹ column 3, were obtained from our wavelengths as given in Table I by the conversion formula:

$$E = \frac{12372.44 \pm 0.16}{\lambda (x-units)} \text{ kev.}$$
(1)

There is a slight systematic discrepancy between the energies given in reference 9 and those obtained from the present wavelength measurements. The present

⁸ E. Ingelstam, Nova Acta Regiae Soc. Sci. Upsaliensis 4, No. 5 (1936).

⁹Y. Cauchois, J. phys. radium 16, 253 (1955); 13, 113 (1952). ¹⁰ P. Sakellaraides, Compt. rend. 236, 1767 (1954). ¹¹ Cohen, DuMond, Layton, and Rollett, Revs. Modern Phys. 27, 363 (1955).

					were identified	i by compar	ing the
Element	Line	Wavelength in Siegbahn x-units	Previous n (x-1	neasurements units)	transition ene energies given doublets but t	rgies which in referenc hev cannot	could e 9. Th be reso
Neodymium	$K_{m eta 3} \ K_{m eta 1}$	293.45 ± 0.05 292.71 ± 0.05	293.51ª 292.75ª	293.410 ^ь 292.683 ^ь			Energy puted fi
Samarium	$K_{\beta 3}$	273.20 ± 0.04	273.25ª		Element	Line	mea
	$K_{\beta 1}$	272.45 ± 0.04	272.50ª		Neodymium	$K_{\beta 3}$	42
	$K_{\beta 5}$ $KO_{11}O_{111}$	270.36 ± 0.02 264.37 ± 0.02	•••			$K_{\beta 1}^{\mu 0}$	42
~		000 44 - 0.04	000.44	000 (4)	Samarium	$K_{\beta 3}$	45
Gadolinium	$K_{\alpha 2}$	292.44 ± 0.04	292.41 ^a	292.01°		$K_{\beta 1}$	45
	$\kappa_{\alpha 1}$	287.75 ± 0.04 254.81 ± 0.03	287.73	201.020		$K_{\beta 5}$	45
	K_{B1}	254.07 ± 0.03	253.94ª			VOUO III	40
	K_{85}	252.23 ± 0.02			Gadolinium	$K_{\alpha 2}$	42
	KO ₁₁ O ₁₁₁	246.36 ± 0.02	•••			$K_{\alpha 1}$	43
						$K_{\beta 3}$	48
Terbium	$K_{\alpha 2}$	$282.84 {\pm} 0.04$	282.94ª	282.86°		$K_{\beta 1}$	48
	$K_{\alpha 1}$	278.14 ± 0.04	278.19ª	278.20°		$K_{\beta 5}$	49
	$K_{\beta 3}$	240.32 ± 0.03 245.57 ± 0.03	240.29ª			KOnOm	50
	K_{β_1}	245.57 ± 0.03 238.00 ± 0.02	245.514			Ronom	
	Ronom	200.09 - 0.02			Terbium	$K_{\alpha 2}$	43
Dysprosium	K	273.66 ± 0.04	273 64ª	273 750		$K_{\alpha 1}$	44
	11.02	(273.68 ± 0.04)	210.01	210.15		$K_{\beta 3}$	51
	$K_{\alpha 1}$	268.94 ± 0.03	268.87ª	269.03°		K_{β_1}	. 51
		(268.97 ± 0.03)				Ronom	
	$K_{\beta 3}$	238.11 ± 0.02	237.87ª		Dysprosium	$K_{\alpha 2}$	45
	Ka	(238.14 ± 0.02)	027 108			$K_{\alpha 1}$	46
	Λ β1	(237.38 ± 0.02)	257.10-			$K_{\beta 3}$	51
	Kas	235.69 ± 0.02				$K_{\beta 1}$	52
	$KO_{11}O_{111}$	230.08 ± 0.02	•••			.1\$5	52
	77	0(1.01 + 0.02	0.000			$KO_{11}O_{111}$	53
Hoimium	$K_{\alpha 2}$	264.91 ± 0.03 260.10 ± 0.03	264.99		Holmium	Kan	46
	$K_{\alpha \alpha}$	200.19 ± 0.03 230 35 ± 0.02	200.30		Hommuni	$K_{\alpha 1}$	47
		229.64 ± 0.02	• • • •			$K_{\beta 3}$	53
	$K_{\beta 5}^{\beta 1}$	228.08 ± 0.02	· . • • •			$K_{\beta 1}$	53
	KO ₁₁ O ₁₁₁	222.59 ± 0.02	•••			$K_{\beta 5}$	54
Erbium	K	25655 ± 0.03	256 72≞	256 64		$KO_{11}O_{111}$	55
	$\widetilde{K}_{\alpha 1}$	251.82 ± 0.03	251.99ª	251.97°	Frhium	K	48
	$K_{\beta 3}$	222.95 ± 0.02	223.00ª		Libium	$K_{\alpha 1}$	49
	$K_{\beta 1}$	222.20 ± 0.02	222.15ª			$K_{\beta 3}$	55
	$K_{\beta 5}$	220.79 ± 0.02	•••			$K_{\beta 1}$	55.
	MOHOIH	213.37 ± 0.02				$K_{\beta 5}$	56
Thulium	$K_{\alpha 2}$	248.59 ± 0.03	248.61ª			$KO_{11}O_{111}$	57.
	$K_{\alpha 1}$	243.79 ± 0.03	243.87*		Thulium	Ko	40
		215.91 ± 0.02 215.11 ± 0.02	215.58" 214.87a		Inditum	$K_{\alpha 1}$	50
	Kas	213.60 ± 0.02				$K_{\beta 3}$	57
	$\widetilde{KO}_{11}O_{111}$	208.48 ± 0.02				$K_{\beta 1}$	57.
						$K_{\beta 5}$	57.
Ytterbium	$K_{\alpha 2}$	240.94 ± 0.03 236 16 ± 0.03	240.99ª 236.22ª	240.98°		KO11O111	59.
	Kai	209.57 ± 0.02	209.16ª	200.20	Vittorhium	V	51
	$\widetilde{K}_{B1}^{\mu\nu}$	208.55 ± 0.02	208.34ª		rtterblum	$K_{\alpha 2}$	52
	$K_{\beta 5}$	206.96 ± 0.02	•••			Kas	59
	$KO_{11}O_{111}$	201.84 ± 0.02	•••			$K_{\beta 1}$	59
Lutetium		000 50 0 0 00	000 70			$K_{\beta 5}$	59.
	$K_{\alpha 2} K_{\alpha 1}$	233.59 ± 0.03 228.81 ± 0.03	233.58ª 228.82ª			KO11O111	61.
	$K_{\beta 3}$	202.09 ± 0.02	202.52ª		Tutetium	Kc	50
	<u>Κ</u> β5	200.42 ± 0.02 105.48 + 0.02	•••		Luccium	$K_{\alpha 1}^{\Lambda \alpha 2}$	54
	TOTOTI	190,1010002			1	TZ I	61

TABLE I. The x-ray wavelengths of the rare earth elements are shown. The present values are compared with previous direct measurements of the wavelengths of these x-rays. -----

Reference 6.
Reference 8.
A. Leide, Compt. rend. 180, 1203 (1925); and dissertation, Lund, 1925 (unpublished).

TABLE II. The x-ray energies obtained from the wavelengths in Table I are compared with the transition energies computed from the tables given in reference 9. The weak lines ($K_{\beta\beta}$ and $KO_{11}O_{111}$) were identified by comparing their energies with all the possible transition energies which could be calculated from the level energies given in reference 9. The $K_{\beta\beta}$ and $KO_{11}O_{111}$ lines are doublets but they cannot be resolved by the present method

292.083*	Element	Line	Energy (in kev) com- puted from wavelength measurements	Energy (kev) computed from reference 9
	No.	V.	42.16 + 0.01	42.150
	Neodymium	$\kappa_{\beta 3}$	42.10 ± 0.01	42.139
		$K_{\beta 1}$	42.27 ± 0.01	42.200
	Samarium	K_{B3}	45.29 ± 0.01	45.293
292.61°		$K_{\beta 1}$	45.41 ± 0.01	45.415
287.82°		$K_{\beta 5}$	45.73 ± 0.01	$45.727 (K - M_{IV})$
		KOIIOIII	46.80 ± 0.01	46.813
	Gadolinium	$K_{\alpha 2}$	42.30 ± 0.01	42.284
		$K_{\alpha 1}$	43.00 ± 0.01	42.971
		$K_{\beta 3}$	48.50 ± 0.01	48.522
2.86°		$\kappa_{\beta 1}$	48.70 ± 0.01	48.007
8.20°		Λβ5	49.05±0.01	$40.990 (K - M_{IV})$ $40.027 (K - M_{IV})$
		K00	50.22 ± 0.01	49.027 (K - MV)
		Ronom	30.22 - 0.01	50.190
	Terbium	$K_{\alpha 2}$	43.74 ± 0.01	43.731
750		$K_{\alpha 1}$	44.48 ± 0.01	44.468
5.13		$K_{\beta 3}$	51.96 ± 0.01	51.911
0 020		$K_{\beta 1}$	52.12 ± 0.01	52.094
7.03°		$KO_{II}O_{III}$	51.97 ± 0.01	51.954
	Dumparti	K.	45 21 + 0.01	45 180
	Dysprosium		43.21±0.01	45.109
		$K_{\alpha \alpha}$	51 06-10 01	51 011
		Ken	52.12 ± 0.01	52.094
		Kar	52.49 + 0.01	$52.436 (K - M_{TT})$
				$52.474 (K - M_v)$
		$KO_{II}O_{III}$	53.77 ± 0.01	53.743
	Holmium	K	46 700 01	46 681
	riomium	K_{a2}	47.55 ± 0.01	47 526
			5371 ± 0.01	53 668
	5	Kai	53.88 ± 0.01	53.849
		\widetilde{K}_{85}	54.25 ± 0.01	54.205 $(K - M_{\rm IV})$
	•			54.245 $(K - M_V)$
6 4 ¢		$KO_{II}O_{III}$	55.58 ± 0.01	55.571
.04° 07¢	E.h.	V	40.02 + 0.01	10 106
.71-	Erbium	$K_{\alpha 2}$	48.23 ± 0.01	48.190
		K _{α1} K _a	49.13 ± 0.01	49.100
		$K_{\alpha 1}$	55.49±0.01	55 776
		Γβ1 Kor	56.00 ± 0.01	56.005 (K - M)
		11 p 5	30.07±0.01	$56.049 (K - M_{T})$
		KO11O111	57.45 ± 0.01	57.425
	(7)] 1	77	10 77 1 0 01	40 750
	Inulium	$K_{\alpha 2}$	49.77 ± 0.01	49./38
		$K_{\alpha 1}$	50.75 ± 0.01	50.125 57 970
		K_{α}	57.50 ± 0.01 57.52 \pm 0.01	51.219 57 181
		Γβ1 Ker	57.52 ± 0.01 57.02 ± 0.01	57.404 57.857 (K - M)
		11 p 5	51.54 ±0.01	$57.902 (K - M_{\rm W})$
98° 28°		KO110111	59.35±0.01	59.337
	Ytterbium	$K_{\alpha 2}$	51.35 ± 0.01	51.326
		$K_{\alpha 1}$	52.39 ± 0.01	52.360
		$K_{\beta 3}$	59.04 ± 0.01	59.129
		$K_{\beta 1}$	59.33 ± 0.01	59.352
		$K_{\beta 5}$	59.78 ± 0.01	59.727 $(K - M_{IV})$
		KOmOm	61.30 ± 0.01	$59.774 (K - M_V)$
				#2 0 #0
	Lutetium	$K_{\alpha 2}$	52.97 ± 0.01	52.959
		$K_{\alpha 1}$	54.07 ± 0.01	54.003
		$\kappa_{\beta 3}$	01.22 ± 0.01	01.042 61.667 (V M
_		12.82	01.75至0.01	$61.717 (K - M_{\rm W})$
1 100	.	K0110111	63.29 ± 0.01	63.279
na, 192.	7			



FIG. 2. K-series x-ray spectra of Dy, Ho, and Er. This plate was made by exposing Dy, Ho, and Er targets for 10 ma-hr. The K_{a2} $K_{\alpha 1}$, $K_{\beta 3}$, and $K_{\beta 1}$ lines of each of the elements are visible on the plate.

measurements are all of the order of 10–20 ev larger than those quoted from reference 9 in Table II. One reason for this discrepancy is that the conversion factor between wavelength (x-units) and energy used in reference 9 was apparently 12 370.48 instead of the one given in Eq. (1). This is essentially the conversion factor given by R. T. Birge in his 1942 evaluation of the constants,¹² and it has been long since superseded in later evaluations based on more accurate input data. This would account for roughly 10 ev of the observed discrepancy and would bring the energy values quoted in reference 9 into essential agreement with the present values within their quoted standard deviations. The identification of the weak lines $(K_{\beta 5} \text{ and } KO_{II}O_{III})$ was made on the basis of the comparison shown in Table II.

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¹² R. T. Birge, *Reports on Progress in Physics* (Taylor and Francis, Ltd., London, 1942), Vol. VIII, p. 131. On this page Birge gives the conversion, $(12\ 395.4\pm2.1)\times10^{-8}$ cm abs. volt. Dividing this by 1.00203 to obtain the constant on the Siegbahn scale of x-units, one obtains 12 370.3-kev x-units.



FIG. 1. K-series x-ray spectrum of samarium. This plate was exposed for 100 ma-hr. For a part of the exposure, the lower portion of the plate was shielded with a lead screen. The weak transitions $(K_{\beta5} \text{ and } KO_{II}O_{III})$ are easily seen on the plate.



FIG. 2. K-series x-ray spectra of Dy, Ho, and Er. This plate was made by exposing Dy, Ho, and Er targets for 10 ma-hr. The K_{α^2} K_{α^1} , K_{β^3} , and K_{β^1} lines of each of the elements are visible on the plate.