NOTIZEN

Precise Energies of K-Röntgen-Lines of Tm, Th, U and Pu

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(Z. Naturforsch. **31 a**, 102–104 [1976]; received December 5, 1975)

The energies of the $K\alpha_1$- and $K\alpha_2$-Röntgen lines of Tm, Th, U and Pu have been measured with high accuracy relative to the 411.794 keV γ-line from a gold source. In addition, the energies of a few $K\beta$ lines have been determined.

1. Introduction

Accurate energies of K-Röntgen-lines of heavy elements are of interest for a comparison with a theory [1] which allows precise calculations of atomic electron binding energies. They are also needed for the calibration of Röntgen- and Ge(Li)-spectrometers.

In practice the K-Röntgen-line energies are calculated from the atomic level energies listed in standard tables such as those given by Hagström, Nordling and Siegbahn [2] or by Lederer, Hollander and Perlman [3] or they are calculated from the energies recommended by Bearden and Burr [4]. Inspection of such tables shows that differences up to 5×10^{-4} in relative energy (or 58 eV at 121.8 keV) still exist for the energy of the 1s state in Pu.

Therefore, the $K\alpha_1$- and $K\alpha_2$-line energies of a few heavy elements have been determined with very high accuracy with our interferometric curved crystal diffractometer [5].

2. Experimental Method

The measurements were performed with the aid of the curved crystal spectrometer which has been described elsewhere [5]. For the Tm measurement a very thin source was fabricated out of 9 mg of Yb_2O_3 enriched in ^{168}Yb to about 20%. The U-measurement was carried out with the use of a similar probe made out of 25 mg of metallic thorium powder. For the measurement of the Pu-lines a metallic uranium foil with a weight of 91 mg and depleted with respect to ^{235}U was used as source. All of these sources were activated in the reactor Merlin of the Kernforschungsanlage Jülich. The thorium lines were measured twice: a) during the U-measurement (fluorescence due to the ^{233}Pa decay) and b) during the irradiation of a small sample of 1.5 mg of thorium metal powder located between two gold sources of a total activity of about 50 Ci. In this latter case, a ^{169}Yb source was used as standard. The U-fluorescence measurement was performed in the same manner but with a gold source as standard because of the proximity of the 93 keV line of ^{169}Tm. During the other measurements either a gold source or a ^{169}Yb source provided the reference line. All measurements were performed in the manner outlined in detail previously [5, 6]. The resolution ΔE(FWHM) at the diffractometer was $\Delta E = 50$ eV during the Tm-$K\alpha$-measurement which could only be performed in the first order of reflection (see Fig. 1) because of the limited angular range of the instrument. It was $\Delta E \approx 33$ eV for the Tm $K\beta_1$ line and $\Delta E \approx 63$ eV during the $K\alpha$-measurement of Pu ($\Delta E \sim E^2$).

3. Results and Discussion

The results of the present experiment are listed in Table 1. The relative energies are based on the 411 keV gold standard. For an estimate of the influence of the particular choise of the standard ener-

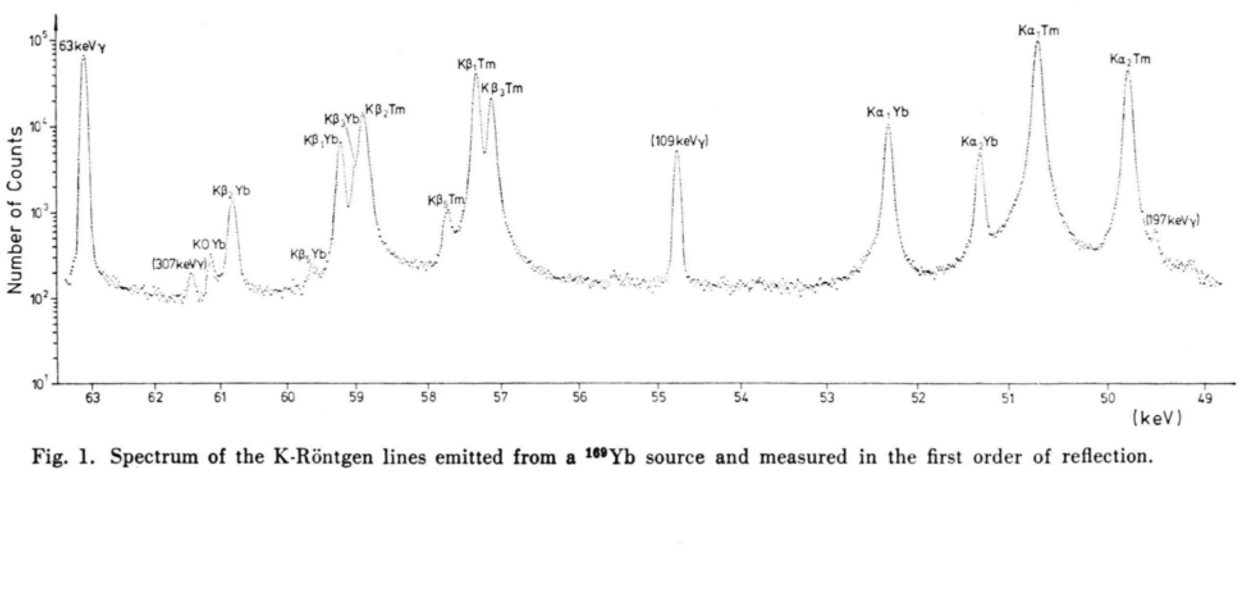

Fig. 1. Spectrum of the K-Röntgen lines emitted from a ^{169}Yb source and measured in the first order of reflection.

Table 1. Energies $E_{relative}$ and absolute energy errors dE of K-Röntgen lines of Tm, Th, U and Pu excited during the decay of ^{169}Yb, fluorescence, the decay of ^{233}Pa and of ^{239}Np respectively and measured relative to the 411.794 keV γ-ray standard from the decay of ^{198}Au. The error of the 411.794 keV line, which is given as ± 7 eV has therefore not been included in the results listed in column 4 of the table. For comparison with previous data, level-energy diffrences are tabulated in the columns 5, 6 and 7.

Element	Radioactive Source	Röntgen Line	$E_{relative} \pm dE$/eV		Hagström et al.	Lederer et al.	Bearden and Burr E/eV $\pm dE$/eV		E/eV
Tm	^{169}Yb → ^{169}Tm	Kα_1	50,740.52	0.09	50,742	50,742	50,741.6	0.6	50,741.35
	in	Kα_2	49,771.73	0.12	49,772	49,772	49,772.7	0.6	49,772.54
	20% $^{168}Yb_2O_3$	Kβ_1	57,507.68	0.15	57,502	57,505	57,505.1	1.2	57,508.62
Th	^{233}Pa in	Kα_1	93,346.0	0.6	93,349	93,351	93,350.6	1.0	93,347.5
	Th metal	Kα_2	89,955.5	0.7	89,957	89,958	89,957.7	1.0	89,957.0
	fluorescence	Kα_1	93,345.8	0.6	93,349	93,351	93,350.6	1.0	93,347.3
	Th metal	Kα_2	89,954.9	0.7	89,957	89,958	89,957.7	1.0	89,956.4
		Kβ_3	104,817.7	1.6	104,818	104,820	104,820.5	1.0	104,819.4
U	^{233}Pa → ^{233}U	Kα_1	98,431.9	0.6	98,436	98,438	98,439.8	1.6	98,433.5
	in	Kα_2	94,651.0	0.7	94,656	94,658	94,658.5	1.6	94,652.6
	Th metal								
	fluorescence	Kα_1	98,431.2	0.8	98,436	98,438	98,439.8	1.6	98,432.8
	U metal	Kα_2	94,650.1	0.8	94,656	94,658	94,658.5	1.6	94,651.7
Pu	^{239}Np → ^{239}Pu	Kα_1	103,732.1	0.6	103,707	103,761	103,761	44	103,733.8
	in	Kα_2	99,521.3	1.2	99,497	99,552	99,552	44	99,522.9
	U metal								
W	fluorescence	Kα_1	59,318.04	0.20	59,320	59,320	59,318.2	0.4	59,319.01

gy on the Röntgen-line energies, the table includes the relative energy of the Kα_1 line of tungsten. Our result of 59,318.04 $\pm$ 0.20 has been obtained in the following way:

relative energies (based on the gold standard) of Kα_1- lines emitted from different sources had previously been measured [7] by the author.

The results were the following:

59,317.97 $\pm$ 0.30 (^{187}W source obtained through neutron activation of W),

59,318.0 $\pm$ 0.6 (W inactive, fluorescence induced by a ^{198}Au source)

59,318.14 $\pm$ 0.30 (W inactive, fluorescence induced by a ^{153}Sm source).

They lead to a weighted mean of 59,318.04 $\pm$ 0.20. If one adopts Cohen and Taylor's 1973 value [8] for the Kα_1 energy of tungsten: 59,319.01 eV $\pm$ 0.33 eV one arrives at two conclusions:

1) Our relative energy of 59,318.04 $\pm$ 0.20 based on the 411 eV line leads to an energy of 411,800.7 eV $\pm$ 2.7 eV for the gold line.
2) All of our entries $E_{relative}$ must be increased by 1.63×10^{-5}. We then obtain the obsolute energies E (in eV) based on Cohen and Taylor's value and listed in the last column of Table 1.

Inspection of Table 1 shows that with respect to the energies for the Kα_1 line of tungsten reasonable agreement exists between our results for the Kα_1 and Kα_2 lines of Tm and the data tabulated by Hagström et al. [2], Lederer et al. [3] and Bearden and Burr [4], although the latter energies appear to be systematically larger by about 1 eV. The energy of our Kβ_1 line of Tm is, however, significantly larger than the other values.

Since a comparison of line-energies based on different standards is always difficult, we choose to compare line-energy differences. In this case the differences of the standard energies have relatively little or negligible influence on the numbers to be compared. One can immediately realize that our figure agrees well with the result of Bearden and Burr [4] and fairly well with the identical results of Hagström et al. [2] and Lederer et al. [3] regarding $E(L_{II}) - E(L_{III})$.

For the Kβ_1 line of Tm we obtain $E(K\beta_1) - E(K\alpha_2) = 6{,}767.16 \pm 0.17$ eV which does not agree with the differences of 6.760[2], 6,763[3] and 6.763 $\pm$ 1.4 eV of the tabulated level energies. In order to understand this discrepancy we have studied the energy systematics of the $E(L_{III}) - E(M_{III})$ distance which should equal the difference $E(K\beta_1) - E(K\alpha_2)$ in the absence of line assymmetries. It must be emphasized that our K-Röntgen energies listed in Table 1 are those of the quanta emitted by the sources characterized in the second column of that table. For other sources shifts [9] may arise which may even exceed our quoted errors. It is obvious that this energy distance should increase monotonously with Z. Testing the tabulated data [2–4] in this respect should, therefore, provide insight into the magnitude of the statistical uncertainties and reveal possible errors of individual distances.

Table 2. Comparison between the K-Röntgen line-energy differences as obtained in the present work and the tabulated level energy differences. The units are eV.

Element	Levels	Bearden and Burr	Lederer eal al.	Hagström et al.	this work
Tm	$E(L_{II})-E(L_{III})$	$1{,}031.1\pm0.8$	1.030	1.030	$1{,}031.21\pm0.15$
Th	$E(L_{II})-E(L_{III})$	$3{,}392.9\pm0.5$	3.393	3.392	$3{,}390.7\ \pm1.7$
	$E(L_{II})-E(M_{II})$	$14{,}862.8\pm0.6$	14.862	14.861	$14{,}862.5\ \pm1.7$
U	$E(L_{II})-E(L_{III})$	$3{,}781.3\pm0.4$	3.780	3.780	$3{,}781.0\ \pm0.8$
Pu	$E(L_{II})-E(L_{III})$	$4{,}209.4\pm0.9$	4.209	4.210	$4{,}210.8\ \pm1.4$

In the region $60\leqq Z\leqq 78$ the distance between the L_{III} and M_{III} levels can be described very well with the aid of the empirical relation:

$E(L_{III})-E(M_{III})\approx\tilde{E}=$
$6767\ \text{eV}+(Z-69)\times222.5\ \text{eV}+(Z-69)^2\times1.8\ \text{eV}$.

In the lower part of Fig. 2 we have plotted the differences $[E(L_{III})-E(M_{III})]-\tilde{E}$. The figure clearly shows that the tabulated [2–4] level distance $E(L_{III})-E(M_{III})$ is not consistent with the expectation. The result of the present experiment is, however, in full agreement with the systematics. The discrepancy is due to the large errors and inconsistence of the tabulated M_{III} level energies [2–4] for $Z=69$. This is demonstrated in the upper part of Fig. 2 where again a smooth function is expected [with $\tilde{e}=70\ \text{eV}+(Z-69)\times2\ \text{eV}$].

Our energies for the thorium lines are systematically somewhat smaller than the tabulated level distances. This is also true for the differences of $E(K\alpha_1)-E(K\alpha_2)$ and of $E(L_{II})-E(L_{III})$. In case of the distances $E(K\beta_3)-E(K\alpha_2)$ and $E(L_{II})-E(M_{II})$ very good agreement is observed with the tabulated data (see Table 2).

The $K\alpha_1$ and $K\alpha_2$ Röntgen-line energies of U of $E_{\text{relative}}=98{,}431.6\pm0.5\ \text{eV}$ and $94{,}650.6\pm0.6\ \text{eV}$ are considerably smaller than the tabulated level-energy differences. Inspection of Table 2 shows very good agreement between the $L_{II}-L_{III}$ level energy differences and our difference $E(K\alpha_1)-E(K\alpha_2)$. This indicates that the tabulated K-shell energies are to large by $5-8$ eV.

In case of plutonium, the tabulated [2, 3] K-shell energies differ by as much as 58 eV. The uncertainty of the result by Bearden and Burr [4] is 44 eV. Our K-line energies are larger than the energies of Hagström et al. [2] and smaller than those of Lederer et al. [3]. They agree within the 44 eV error with the energies of Bearden and Burr. In view of our much higher accuracy we have again compared the line-energy differences with the tabulated [2–4] level-energy distances. Table 2 shows agreement for the $L_{II}-L_{III}$ spacing.

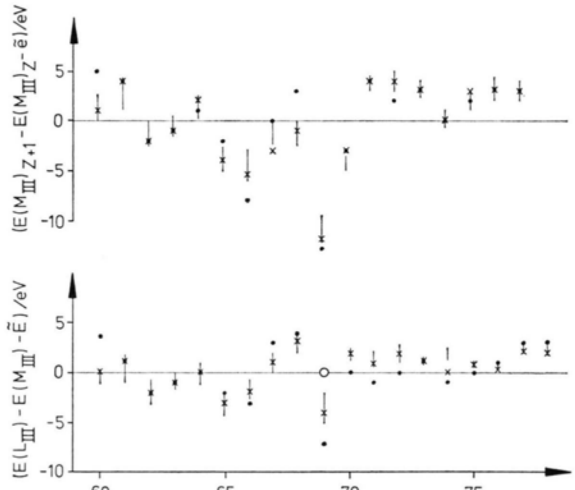

Fig. 2. Deviations of the tabulated distances $E(L_{III})-E(M_{III})$ from the approximate energy difference $\tilde{E}$ and of the tabulated differences $E(M_{III})_{Z+1}-E(M_{III})_Z$ from the approximate energy difference $\tilde{e}$ for $60\leqq Z\leqq78$. The data of Hagström et al. [2] are marked ●, those of Lederer et al. [3] are given by × and the values of Bearden and Burr [4] by the vertical line whose length corresponds with the quoted error. The open circle is the result of the present experiment.

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