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June 1964

DETERMINATION OF THE ATOMIC ELECTRON BINDING ENERGIES IN ELEMENT 97

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The atomic electron binding energy is defined as the energy required to raise an electron from a bound atomic state to the lowest continuum state. Knowledge of the values of atomic binding energies is of importance in nuclear spectroscopy because of their application to the determination of nuclear transition energies from internal conversion electron data.

Two general methods have been used for the determination of electron binding energies. The first method utilizes a combination of X-ray absorption and emission spectroscopy; several tables of binding energies so obtained are in use.^{1,2} In recent years an electron spectroscopic method has been developed by which the energies of photoelectrons ejected from suitable targets by X-radiation of known wave length are measured by means of a high-precision electron spectrometer.³ A table of binding energies incorporating these data has recently been prepared by Hagström, Nordling, and Siegbahn.⁴

The use of either method requires that the target material contain a macro quantity of the element under study. As a consequence, the very heavy elements, not generally available in large quantities, have received inadequate study, and in fact most of the quoted binding energies for heavy elements have been extrapolations from lower atomic number. Uranium ($Z = 92$) is the heaviest element to have been studied with the photoelectron method;³ with X-rays, americium ($Z = 95$) has received some study.⁵ As pointed out by Hagström et al.,⁴ the presently quoted electron binding energies obtained

by extrapolation may be in error by more than 100 eV. Thus it is desirable to have measurements, wherever possible, of the binding energies of high-Z elements.

In some cases knowledge of certain properties of a nuclear level scheme makes possible the determination of the atomic binding energies in a manner independent of X-ray spectroscopy. Such a case is the simple cascade-crossover situation illustrated by the partial level scheme of Fig. 1. In this scheme, the absolute transition energy of transition A may be found from the energy difference between any two like (i.e., same subshell) conversion lines from transitions B and C. From the absolute energy of transition A one can compute the binding energies of the various subshells by subtracting the measured energies of the corresponding subshell conversion electron lines from the transition energy.

In the course of a detailed study of the decay of $\text{Es}^{253,6}$ we have had occasion to measure with high accuracy the energies of a number of internal conversion lines in ${}_{97}\text{Bk}^{249}$, and from these data we can calculate the atomic binding energies of berkelium ($Z = 97$). In the level scheme of Bk^{249} (shown partially in Fig. 2) several cascade-crossover sequences are prominent and can be used for this purpose. These are:

$$E_{\gamma} (51.9) = E_i (93.7) - E_i (41.8)$$

$$E_{\gamma} (43.0) = E_i (73.8) - E_i (30.8)$$

where i represents internal conversion in subshell i .

The internal conversion spectrum was studied with the Berkeley 50-cm $\pi\sqrt{2}$ iron-free spectrometer.^{7,8} This instrument is programmed to scan automatically with pre-selected current-step intervals and counting times, and

the relevant output data for each current setting were printed out by an IBM output-writer. During these measurements, it was operated on a 24-hour, 7-day per week basis. Absolute current measurements were made with use of a Leeds and Northrup Type K-2/ potentiometer, by measuring the IR drop across a 0.01 Ω precision series-resistor maintained in a constant ($\pm 0.01^\circ \text{C}$) temperature bath. At the time these measurements were made the current stability of the spectrometer power supply was about 3:10,⁵ and the current could be measured with comparable precision. Other limitations on the accuracy of the B_0 determinations came from the uncertainties in determining the line positions and from the spectrometer calibration error.

Calibration of the spectrometer was made with reference to the K line of the 662-keV transition in Ba^{137} (Cs^{137} source) which has been measured relative to the internal conversion lines of the 412-keV transition in Hg^{198} .⁹ The Hg^{198} transition energy has recently been measured with respect to annihilation radiation with high accuracy.¹⁰ When analyzed with use of the 1963 values of the fundamental constants¹¹ the Hg^{198} transition energy is 411.795 ± 0.009 keV,¹² the Ba^{137} transition energy is 661.636 ± 0.053 keV and the K 662 line has a momentum value of 3381.28 ± 0.20 gauss cm. This B_0 value is ~ 6 parts in 10^5 higher than the value reported earlier⁹ which was based on the 1955 constants. For making momentum calibrations the "standard" source and "unknown" source were alternately moved into the electron-optical source position by means of a suitably designed holder that accommodates both sources and allows their interchange without removing the source holder from the instrument.

A portion of the measured conversion spectrum is shown in Fig. 3. The data used for calculation of the absolute transition energies of the 51.9- and 43.0-keV transitions are shown in Table I. The following

contributions to the errors in the absolute values of the subshell-conversion-line energies were considered:

- 1) Irreproducibility of source holder during interchange of unknown with standard sources, $1:10^4$, as determined from independent experiments.
- 2) Uncertainty in establishing the peak position current of the conversion lines. This error was taken as $1.5:10^4$ for all lines. The peak positions were determined by extrapolating the locus of midpoints of the conversion line to its intersection with the top of the line (see Fig. 3).
- 3) Potentiometer inaccuracy, estimated to be $5:10^5$.
- 4) Uncertainty in the calibration constant; estimated to be $8:10^5$.

The first three errors were assumed to be statistical while the fourth was assumed to be systematic. In Table I only the statistical errors were considered and therefore the errors in the Difference column were added quadratically.

From the absolute transition energies given in Table I, the various subshell binding energies were calculated from the measured energies of the individual conversion lines of these transitions. The results are given in Table II.

The calibration error is included only in the last column and is added linearly. All of the other errors in Table II were added quadratically.

It is not possible to calculate the K-binding energy in the same manner as the others because the transition energies used (51.9- and 43.0-keV) are lower than the K-edge and do not produce K-lines. Therefore, use was made of the carefully measured energy difference between the K and L_I lines of the 389.2-keV transition, which is very prominent in the Es^{253} decay. These

results are also included in Table II. It is interesting to note that the absolute value of the K-binding energy so obtained is higher by 150 eV and ~ 300 eV than the extrapolated values quoted by Hagström et al.,⁴ and by Hyde,² respectively.

Table I. Determination of absolute transition energies of 51.9- and 43.0-keV transitions from Es^{253} decay.

$$A. E_{\gamma}(51.9) = E_1(93.8) - E_1(41.8)$$

Subshell i	$E_1(93.8)$	$E_1(41.8)$	Difference
L_{II}	69.366 ± 0.042	17.404 ± 0.010	51.962 ± 0.043
L_{III}	74.302 ± 0.044	22.340 ± 0.013	51.962 ± 0.045
M_{II}	87.585 ± 0.053	35.638 ± 0.021	51.947 ± 0.056
M_{III}	88.757 ± 0.053	36.807 ± 0.022	51.950 ± 0.056
			$E_{\gamma} = 51.956 \pm 0.025$

$$B. E_{\gamma}(43.0) = E_1(73.8) - E_1(30.8)$$

Subshell i	$E_1(73.8)$	$E_1(30.8)$	Difference
M_{II}	67.657 ± 0.040	24.688 ± 0.015	42.969 ± 0.042
M_{III}	68.839 ± 0.041	25.851 ± 0.016	42.988 ± 0.043
			$E_{\gamma} = 42.979 \pm 0.030$

Table II. Atomic electron binding energies in element 97 (berkelium).

Shell	Transition Energy (keV)	Conversion Line Energy (keV)	Difference (keV)	Selected Value
L _I	51.956 ± 0.025	26.682 ± 0.021	25.274 ± 0.033	L _I 25.275 ± 0.026
	42.979 ± 0.030	17.704 ± 0.014	25.275 ± 0.033	
L _{III}	51.956	27.574 ± 0.022	24.382 ± 0.034	L _{III} 24.385 ± 0.026
	42.979	18.592 ± 0.015	24.387 ± 0.033	
L _{III}	51.956	32.506 ± 0.026	19.450 ± 0.036	L _{III} 19.452 ± 0.030
	42.979	23.525 ± 0.019	19.454 ± 0.040	
M _I	51.956	45.399 ± 0.036	6.557 ± 0.044	M _I 6.556 ± 0.031
	42.979	36.424 ± 0.029	6.555 ± 0.041	
M _{II}	51.956	45.809 ± 0.037	6.147 ± 0.045	M _{II} 6.147 ± 0.046
M _{III}	51.956	46.979 ± 0.037	4.977 ± 0.045	M _{III} 4.977 ± 0.046
N _I	51.956	50.193 ± 0.040	1.763 ± 0.048	N _I 1.755 ± 0.033
	42.979	41.232 ± 0.033	1.747 ± 0.045	
O _I	51.956	51.571 ± 0.041	0.385 ± 0.049	O _I 0.398 ± 0.033
	42.979	42.568 ± 0.034	0.411 ± 0.045	
K	389.2	K-L _I = 106.32 ± 0.05		K 131.59 ± 0.06

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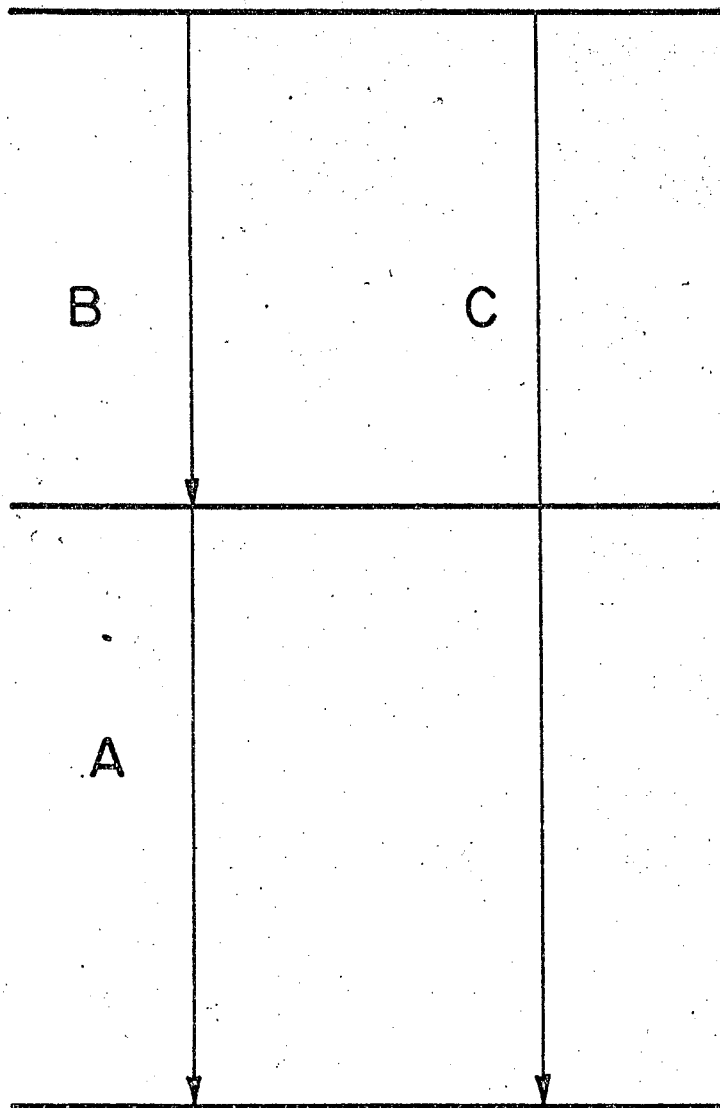
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FIGURE CAPTIONS

Figure 1. Example of cascade-crossover situation in a nuclear level scheme.

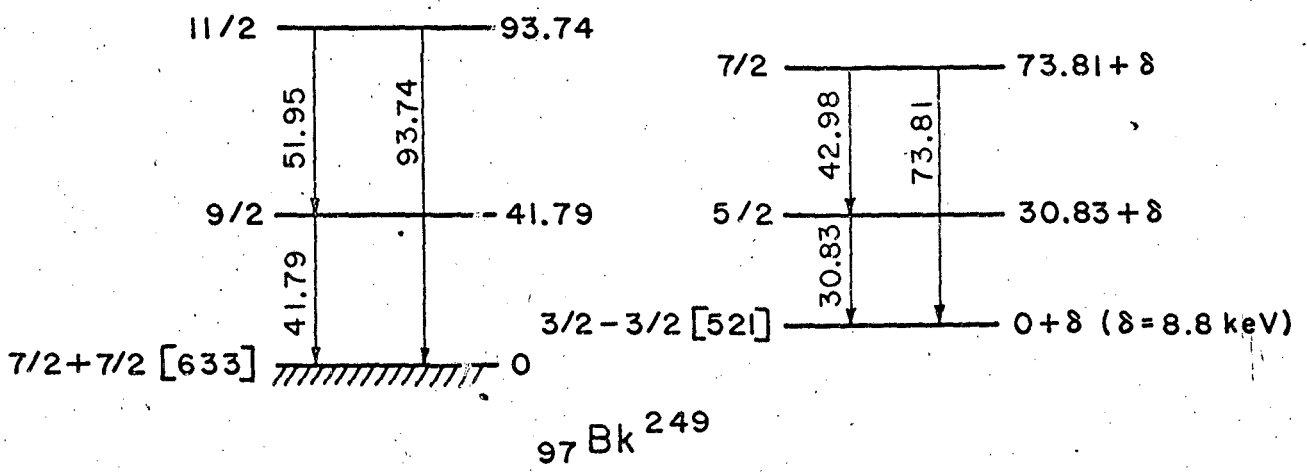
Figure 2. Partial level scheme of ${}_{97}\text{Bk}^{249}$ showing levels used for binding energy determinations.

Figure 3. Portion of the Es^{253} internal conversion spectrum measured with the 50-cm $\pi\sqrt{2}$ iron-free spectrometer.



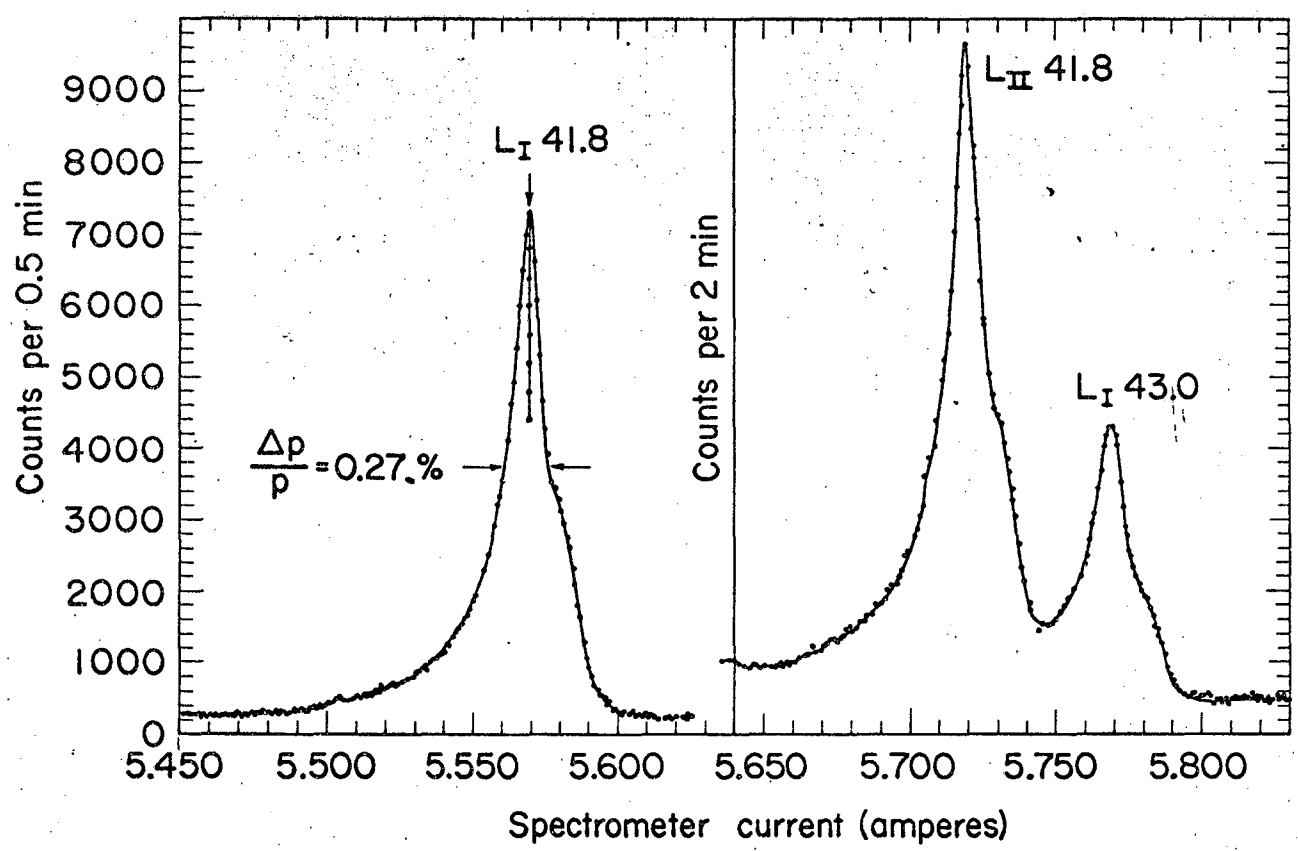
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Fig. 1.



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Fig. 2.



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Fig. 3.

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