$K\alpha_1$ and $K\alpha_2$ X-Ray Energies of Neptunium, Plutonium, and Americium†

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The $K\alpha_1$ and $K\alpha_2$ x-ray energies of Np, Pu, and Am have been measured with a Cauchois-type bent-crystal transmission spectrometer. The energies of these transitions have been determined to an accuracy of 5 eV. The $K\alpha_1 - K\alpha_2$ differences agree within the experimental error with differences obtained from the previously reported L x-ray energies.

INTRODUCTION

The L x-ray spectra of transuranic nuclei have previously been measured to high precision with bent-crystal spectrometers[1-4] and double-crystal spectrometers.[5,6] However, no published measurements have been made of the K x rays from these elements. The main difficulties have been the lack of availability of gram amounts of these elements and a source of sufficient energy and intensity to fluoresce the material.

We have measured the $K\alpha_1$ and $K\alpha_2$ x-ray lines from Np, Pu, and Am by fluorescing 4 g of Np, 51 g of Pu, and 1 g of Am with a 50 Ci Ta^{182} source. The energies of these transuranic x rays were determined to within 5 eV.

EXPERIMENTAL DETAILS

The 2-m Cauchois-type bent-crystal spectrometer used in this experiment has previously been described in detail.[7] It should be mentioned that the detector presently used with the spectrometer is a thin window 2-cm^3 Ge(Li) diode. The Ge(Li) detector significantly improves the signal-to-noise ratio and is very useful for observation of the x-ray or γ-ray pulse-height spectrum.

The source arrangement is new and will be described here in some detail. Figure 1 is a schematic top view of the source holder. The Ta^{182} source consists of a Ta foil contained in a 1×3-in. Al irradiation can. The can is lowered into a Lucite-lined cavity. A $2\times 4\times 8$-in. lead brick covering the top is slotted to permit insertion of the fluorescent foil, which is taped to the Lucite holder. The foil is irradiated by γ rays from the Ta^{182} source. The fluorescent radiation emerges through a converging lead collimator before entering the spectrometer. The fluorescent foils consist of 4 g of Np^{237}, 51 g of Pu^{239}, and 1 g of $Am^{241}O_2$, with each sample sealed in an aluminum can to prevent radioactive contamination.

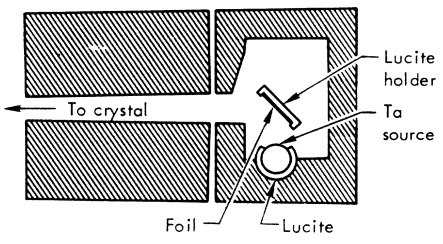

FIG. 1. Source arrangement schematic. Shaded areas are lead.

The x-ray diffraction lines were scanned automatically by a 0.176-mm-wide slit. Two hundred steps of 0.01 mm each were taken across the diffraction peak. The counting time per step varied from 4 to 10 min, according to the intensity of the line being measured. A 400-channel analyzer used in the multiscaler mode recorded the counts at each position. A single-channel analyzer was used to discriminate against any radiation that might differ in energy from the one under investigation.

A least-squares fit was made to the diffraction peaks to determine their position. The function used in this fitting procedure assumes that the observed profile is the fold of the instrumental response with the natural line shape of the x ray. The new shape is in turn folded into the detector slit. The instrumental response is taken as a Gaussian, and the natural line shape of the x ray is assumed to be a Lorentzian.[8] The folding procedure consists of a double integral which was calculated numerically on a CDC-3600 computer. Least-squares fits to the Bi $K\alpha_1$ and Pu $K\alpha_2$ diffraction peaks are shown in Figs. 2 and 3.

The slit position-versus-energy curve was determined from a least-squares fit (see Ref. 7) using the $K\alpha_1$ and $K\alpha_2$ x rays from U, Bi, Pb, and Au, plus the $K\beta_1$ x rays from Th for calibration. The energies reported by Bearden[9] were used as standards.

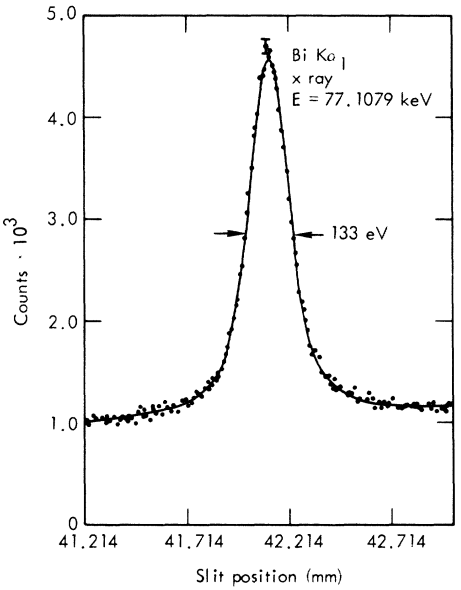

FIG. 2. Profile of the Bi $K\alpha_1$ x ray. The curve is a least-squares computer fit to the data points.

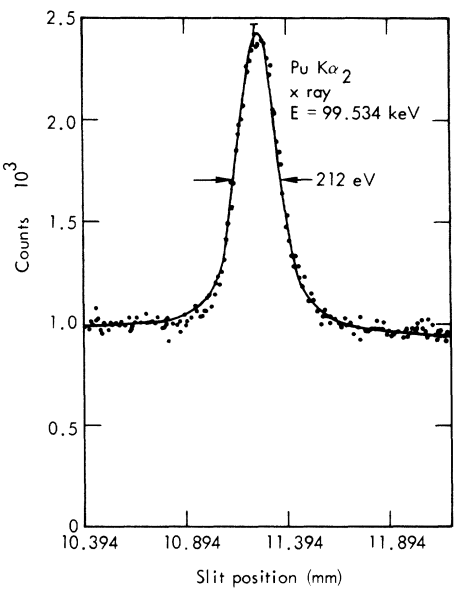

FIG. 3. Profile of the Pu $K\alpha_2$ x ray. The curve is a least-squares computer fit to the data points.

RESULTS AND DISCUSSION

Table I lists the positions and energies of the calibration lines used in the fitting procedure, along with the measured values for Np, Pu, and Am. The errors in the present measurements are the probable errors. The differences between the energies given by Bearden[9] and those calculated from the calibration equation are given in the last column of the table. It should be noted that the 6-eV difference between the calculated U $K\alpha_2$ energy and the U $K\alpha_2$ energy given by Bearden[9] is consistent with the discrepancy in the uranium L_{111}-L_{11} binding energy difference obtained from Bearden's $K\alpha_1$ and $K\alpha_2$ x-ray energies

TABLE I. Data taken to measure the energies of the $K\alpha_1$ and $K\alpha_2$ x rays of Np, Pu, and Am. The first nine lines were used to calibrate the spectrometer. The quoted errors are probable errors. The last column lists the x-ray energy from Ref. 9 minus the energy calculated from the calibration equation.

Line	Energy (keV)	Measured position (mm)	Energy difference (keV)
Th $K\beta_1$	105.609 ±0.008	5.155 ± 0.003	−0.004
U $K\alpha_1$	98.439 ±0.002	12.431 ± 0.001	−0.002
U $K\alpha_2$	94.665 ±0.003	16.715 ± 0.001	0.006
BI $K\alpha_1$	77.1079 ± 0.001	42.115 ± 0.001	−0.001
Pb $K\alpha_1$	74.9694 ± 0.0009	46.029 ± 0.002	−0.0001
Bi $K\alpha_2$	74.8148 ± 0.001	46.321 ± 0.001	0.0002
Pb $K\alpha_2$	72.8042 ± 0.0009	50.222 ± 0.002	−0.002
Au $K\alpha_1$	68.8037 ± 0.0008	58.681 ± 0.001	0.002
Au $K\alpha_2$	66.9895 ± 0.0007	62.842 ± 0.001	−0.0007
Am $K\alpha_1$	106.488 ±0.005	4.334 ± 0.002	
Am $K\alpha_2$	102.044 ±0.004	8.648 ± 0.002	
Pu $K\alpha_1$	103.750 ±0.004	6.948 ± 0.002	
Pu $K\alpha_2$	99.536 ±0.004	11.252 ± 0.002	
Np $K\alpha_1$	101.085 ±0.004	9.629 ± 0.003	
Np $K\alpha_2$	97.095 ±0.004	13.918 ± 0.002	

and that obtained from the more precisely determined $L\beta_1$ and $L\alpha_2$ energies listed by Bearden.

As an added check on our measured values, a comparison was made of the difference in the L_{111} and L_{11} binding energies determined from our $K\alpha_1$ and $K\alpha_2$ x-ray energies and from the previously reported[9] $L\beta_1$ and $L\beta_2$ x-ray energies. These results are shown in Table II. The results agree within our experimental errors of 4 to 5 eV.

Since no previous measurements have been published of the Np, Pu, and Am $K\alpha_1$ and $K\alpha_2$ x-ray energies, the only comparison we can make with our measurements are the values obtained from the appropriate binding energies. Table III lists our measured values, along with the values obtained from the binding energies given by Bearden and Burr[10] and the extrapolated binding energies given by Hagström *et al.*[11] Our values fall between the values of Hagström *et al.*[11] and Bearden for Am and Pu, and 20 to 30 eV above their values for Np. In view of the errors in the binding energies associated with extrapolated values, and the errors of 33, 44, and 55 eV quoted by Bearden and Burr[10] for the K binding energies of Np, Pu, and Am, respectively, our measurements can be considered to be in agreement with, but considerably more precise than, the previous values.

TABLE II. Comparison of the $L_{111}-L_{11}$ binding energy difference calculated from the present $K\alpha_1$ and $K\alpha_2$ x-ray energies and that calculated from the $L\beta_1$ and $L\alpha_2$ x-ray energies listed in Ref. 9.

Element	$K\alpha_1-K\alpha_2$ difference (keV) Present results	$L\beta_1-L\alpha_2$ difference (keV) Reference 9
Am	4.444	4.4401
Pu	4.214	4.2095
Np	3.990	3.9905

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TABLE III. Comparison of the measured $K\alpha_1$ and $K\alpha_2$ x-ray energies for Am, Pu, and Np with the values obtained from the binding energies given in Refs. 10 and 11.

Line	Present results	Hagström *et al.*[a]	Bearden[b]
Am $K\alpha_1$	106.488 ± 0.005	106.427	106.523
Am $K\alpha_2$	102.044 ± 0.004	101.987	102.083
Pu $K\alpha_1$	103.750 ± 0.004	103.707	103.762
Pu $K\alpha_2$	99.536 ± 0.004	99.497	99.552
Np $K\alpha_1$	101.085 ± 0.004	101.054	101.068
Np $K\alpha_2$	97.095 ± 0.004	97.064	97.078

[a]Reference 11.
[b]Reference 10.

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