theory that must be considered. One major source is configuration interaction, and the effects of this (as well as other less important effects) would have to be estimated in Sc II and Ti III before the errors in Ti II could be regarded as fully significant. The variation of the interaction parameter H_2 between the $3d^2$ and the $3d4s$ configuration is fairly well established,[12] and it can be shown that this configuration effect cannot explain the large errors in $y\,^2D$ and $z\,^2P$. However, the interaction parameters R_d and R_e between $3d4p$ and $4s4p$ cannot be estimated on the basis of previous work,[13] and it is not easy to evaluate the perturbation that would result from interaction between these two configurations. For this reason, the $3d4p$ parameters in calculation (2) of Table II might not indicate true behavior. The good agreement obtained would tend to indicate this is not the case and that configuration interaction can be neglected. If this latter assumption were not true, then the Ti II data would be even more perturbed and a linear theory (which includes this method) would be inapplicable.

With regard to the source of the large errors of $y\,^2D$ and $z\,^2P$ in calculation C it should be noted that $y\,^2D$, $z\,^2P$ and $y\,^2F$ have two pairs of interacting levels separated by 450 K in one pair ($J=2\frac{1}{2}$) and by 370 K in the other ($J=1\frac{1}{2}$), so that breakdown of LS-coupling could be appreciable. If the $J=2\frac{1}{2}$ and $J=1\frac{1}{2}$ term assignments were interchanged in these pairs, then the errors of $y\,^2D$, $z\,^2P$, and $y\,^2F$ would be changed from $+468$, -327, and -75 K, respectively, to $+51$, -80, and $+118$ K so that this procedure would remove all significant discrepancies between theory and experiment; the mean deviation of the revised errors would be ± 123 (a new least squares calculation would reduce this still more).

This interchange is probably not warranted, but an appreciable improvement should result if the actual correction were accurately evaluated and applied. It is likely that the classification of the levels given in Volume I of *Atomic Energy Levels* does specify the dominant component. However, the g-values, which would be the most decisive factor in deciding this, are uncertain. A strong interaction between $y\,^2D_{1\frac{1}{2}}$ and $z\,^2P_{1\frac{1}{2}}$ has been noted by H. N. Russell [Mt. Wilson Contract No. 344 (1927) page 30 (unpublished)]. Except for the $z\,^2P_{1\frac{1}{2}}$ g-value (it is not Landé and was probably not assumed), all g-values given for the levels of $y\,^2D$, $z\,^2P$, and $y\,^2F$ terms of Ti II are assumed Landé values (used to calculate other g-values that are given without colons).

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[12] A discussion of this variation and a partial tabulation of values of H_2 is given in Sec. 6 of A. In Sc II and Ti III it is estimated that $H_2=200\pm 20$. The parameters B and C of calculation (2) of Table II were corrected slightly for this effect but no allowance is made for it in calculations A, B, or C of Table I.

[13] The notation for these parameters is that used by N. Rosenzweig [Phys. Rev. **88**, 580 (1952)]. G. Racah [Phys. Rev. **62**, 523 (1942)] has given an evaluation of these parameters in Sc I ($R_d=-3063$ and $R_e=950$) but his calculation neglects polarization effects which are expected to be large. If his interaction parameters are used in Sc II the value of the $z\,^3P-y\,^3P$ interaction element is very large (-4013 K). Since the observed separation of these terms is 9443 K it would be attributed almost entirely to configuration interaction.

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Electromagnetic Spectrum of Am^{241}

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The electromagnetic spectrum of Am^{241} has been studied with a ten-inch bent-crystal spectrometer and a sodium iodide scintillation counter. Twelve gamma rays, sixteen Np L x-rays, and eight Am L x-rays have been measured. The gamma rays in Np have energies of 26.363, 33.199, 43.463, 59.568, 103, 113, 130, 159, 210, 270, 328, and 370 kev. The x-ray energies establish the validity of the Mosley extrapolations in this region of the periodic table. Details of the spectrometer operation and calibration are given.

INTRODUCTION

AMERICIUM-241 is a complex alpha emitter which decays with a 470-year half-life[1] to Np^{237}. A transmission type, bent-crystal spectrometer and a 20-channel scintillation counter have been used to analyze the electromagnetic spectrum emitted during the decay of this isotope. The results are reported herein.

The alpha spectrum has been measured by Asaro *et al.*[2] with a magnetic spectrometer. Six alpha groups were reported with energies of 5.546, 5.535, 5.503, 5.476, 5.433, and 5.379 Mev with relative intensities of 0.23 percent, 0.34 percent, 0.21 percent, 84.2 percent, 13.6 percent, and 1.42 percent, respectively. Later,[3] the 5.546-Mev alpha "peak" was ascribed to scattering from one of the baffles in the spectrometer.

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[1] B. G. Harvey, Phys. Rev. **85**, 482 (1952).

[2] Asaro, Reynolds, and Perlman, Phys. Rev. **87**, 277 (1952).

[3] F. Asaro (unpublished data, September, 1953).

Seaborg *et al.*[4] obtained an energy of 62 kev for the prominent gamma ray by absorption methods with an intensity of about 0.4 gamma ray per alpha particle. Martin,[5] using a gamma-ray pulse analyzer, observed two gamma rays, one at 59 kev with an intensity of 0.4 gamma per alpha particle and another at 100 kev with an intensity of about 10^{-4} gamma per alpha particle. Using a bent-crystal spectrometer similar to the one described in this paper, Barton *et al.*[6] measured the energies of the three prominent L x-ray lines due to internal conversion of the gamma rays in neptunium and found them to agree with the Mosley values extrapolated from thorium and uranium. The alpha-gamma coincident measurements of Prohaska[7] gave 0.32 sixty-kev gamma ray per alpha particle. There were three L-electrons for each M-electron and a total electron coincidence rate of 0.49 electron per alpha particle was obtained. O'Kelley[8] ran the conversion spectrum on the Berkeley beta-ray spectrometer and found the L, M, and N conversion lines for a 59.7-kev gamma ray. A value of 0.28 L-electrons per alpha particle was also quoted. Using a proportional counter, Beling *et al.*[9] observed five lines; 59.7, 26.3, 20.9, 17.3, and 13.5 kev with intensities of 1.0, 0.075, 0.177, 0.66, and 0.42, respectively. They attributed the three lowest lines to the Np L x-rays and by coincidence measurements found 0.40 sixty-kev gamma ray per alpha particle. They[10] also derive upper limits for the conversion coefficients for the 59.7- and 26.3-kev gamma rays of 1.5 and 20, respectively. After modifying the Barton spectrometer, Browne[11] ran the Am^{241} spectrum and found eight Np L x-rays which arise from internal conversion of the gamma rays. In addition, he found six other lines which he attributed to gamma rays with energies of 18.83, 22.20, 26.43, 33.36, 38.00, and 59.78 kev. Newton and Rose[12] showed that the 33.36-kev and 38.00-kev lines found by Browne were due to fluoresence of La which was present in the sample. With proportional counters, they set an upper limit of 0.1 percent per sixty-kev gamma ray for the intensity of a 33-kev gamma ray which was predicted by the level scheme. Using a double-focusing beta spectrometer, Passell[13] found conversion electrons for six gamma rays; 26.3, 33.1, 43.4, 56.4, 59.6, and 99.5 kev. Jaffe[14] found two gamma rays with energies of 26.38 and 59.62 kev using the Barton curved-crystal spectrometer and four gamma rays with energies of 102, 128, 168, and 207 kev using a scintillation spectrometer.

The nuclear level structure of Np^{237} may also be obtained by observing the radiations of the 6.7-day beta emitter U^{237}. Wagner *et al.*[15] found ten gamma rays by the use of scintillation spectrometry and a magnetic lens spectrometer; 27, 43, 59, 102, 165, 207, 269, 334, 370, and 430 kev.

APPARATUS

The work described in this paper has been performed on the ten-inch bent-crystal spectrometer located in a shielded underground room at Argonne National Laboratory. The spectrometer is built along the lines of the transmission spectrometer designed and built at Berkeley by Barton.[6] As originally designed, an elastically bent crystal of quartz was used to diffract the radiation which was detected with a proportional counter. Browne[11] made some important changes on this instrument, the two principal ones being the use of topaz instead of quartz and a scintillation counter instead of the proportional counter. Owing to the increased reflectivity of the topaz over the quartz and the high efficiency of a scintillation counter, Browne was able to improve the transmission of the instrument by a factor of 200. The range of the instrument was 10 kev to 130 kev.

The fundamental design and preliminary assembly of the present spectrometer was carried out by G. Barton assisted by C. A. Prohaska. (The construction drawings were brought from The University of California Radiation Laboratory by Barton.) The author undertook the final assembly and calibration with the guidance and help of Melvin S. Freedman. Although a number of improvements were introduced, basically (see Fig. 1) the instrument is the same as the one modified by Browne.[11] The (303) planes of a topaz crystal 0.008 in.×0.75 in.×1.5 in. (which are inclined at an angle of 29° to the crystal face) elastically bent to a radius of ten inches, are used to diffract the radiation (1).[16] The radiation is detected with a scintillation counter which sets inside of a one-inch lead shield (2). The scintillation counter was designed and constructed by R. Swank and J. Moenich and consists of a rectangular NaI(Tl) crystal (3) 0.25 in.×1.5 in.×2.25 in. in an aluminum can, mounted on the end of a DuMont 6292 phototube (4). A window consisting of a disk of 0.020-in. thick Be with a 0.0002-in. Al reflector next to the crystal has a measured transmission of 95 percent at 14 kev. The detector has a resolution (full width at

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[4] Seaborg, James, and Morgan, *The Transuranium Elements: Research Papers* (McGraw-Hill Book Company, Inc., New York, 1949), Paper No. 22.1, National Nuclear Energy Series, Vol. 14B, Div. IV.

[5] D. F. Martin (unpublished).

[6] G. W. Barton, University of California Radiation Laboratory Report UCRL-670, May, 1950 (unpublished); Barton, Robinson, and Perlman, Phys. Rev. **81**, 208 (1951).

[7] C. A. Prohaska, University of California Radiation Laboratory Report, UCRL-1395, August, 1951 (unpublished).

[8] G. D. O'Kelley, University of California Radiation Laboratory Report, UCRL-1243, 1951 (unpublished).

[9] Beling, Newton, and Rose, Phys. Rev. **86**, 797 (1952).

[10] Beling, Newton, and Rose, Phys. Rev. **86**, 670 (1952).

[11] C. I. Browne, University of California Radiation Laboratory Report UCRL-1764, June, 1952 (unpublished).

[12] J. O. Newton and B. Rose, Phys. Rev. **89**, 1157 (1953).

[13] T. O. Passell, University of California Radiation Laboratory Report, UCRL-2528, March, 1954 (unpublished).

[14] H. Jaffe, University of California Radiation Laboratory Report, UCRL-2537, April, 1954 (unpublished).

[15] Wagner, Freedman, Engelkemeir, and Huizenga, Phys. Rev. **89**, 502 (1953).

[16] Numerals in parentheses refer to the numbered components in Fig. 1.

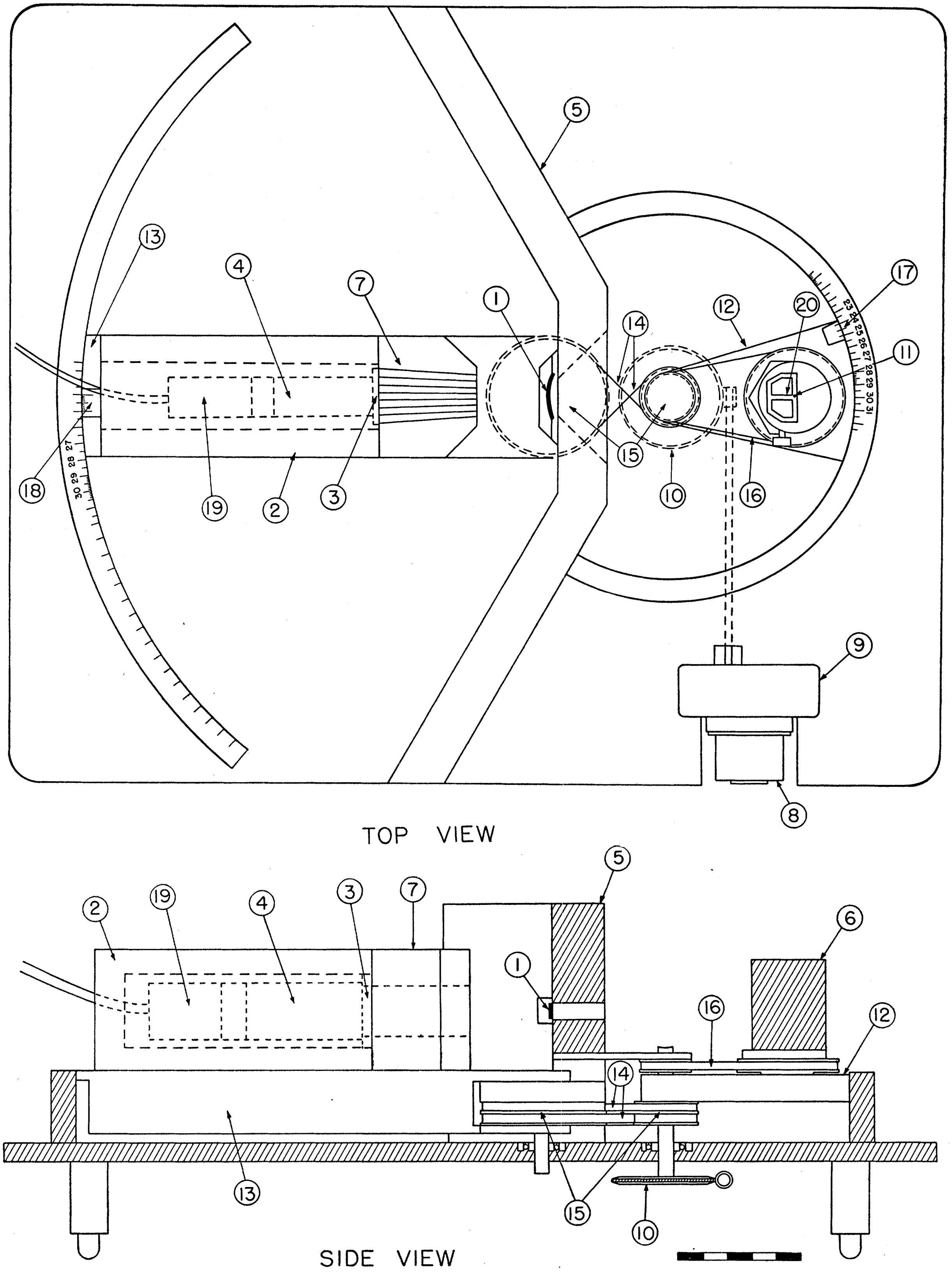

FIG. 1. Schematic diagram of the bent-crystal spectrometer. The source arm (12) and detector arm (13) are shown at 0° Bragg angle for a set of diffracting planes which are perpendicular to the crystal face. e.g., the (310) planes of quartz or the (1000) planes of topaz.

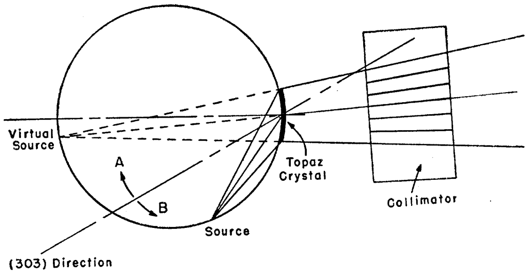

FIG. 2. Schematic diagram illustrating the geometrical optics of the bent-crystal spectrometer.

half-height) of 20 percent at 60 kev and a background (no source in the spectrometer) of about 10 counts/min in the 10-kev region. In order to shield the detector from direct radiation from the source, a stationary 2-in. lead shield (5) is located between the source and the detector. A 1-in. lead shield (6) is placed around the source to keep the radiation level around the spectrometer to a moderate value.

An appreciable gain in transmission over the Browne version was obtained by increasing the topaz crystal dimensions by 30 percent and designing a larger collimator (7) which has an opening sufficient to intercept the entire diffracted beam. The measured transmission of the diffracted beam at 60 kev is 1.7×10^{-6}. The present collimator consist of a truncated pyramidal opening through a 4-in. cube of lead lined with brass plates at the top and bottom in which there are 15 precisely milled grooves, into which fit 0.060-in. thick tungsten sheets. By use of a single collimator with removable baffles it is possible to fulfill the collimation requirements from 8 to $\sim$200 kev.[17] The calculated transmission of the collimator with all 15 tungsten baffles inserted is 44.6 percent in comparison with a measured value of 43.9 percent. Below 30 kev, only 7 baffles are used, to give an increased transmission without a significant increase in the scattered background.

Since the diffracted beam which the collimator intercepts is diverging, the baffles are oriented so that they are parallel to the diverging radiation; thus the collimator will have a "focal point." If the virtual source is located at this "focal point," the diverging rays will pass unhindered through the collimator except for those that are intercepted by the finite thickness of the baffles. It can be seen in Fig. 2 that it is necessary to have the collimator positioned so as to locate its "focal point" at the virtual source. Since the source moves on the circle on which the topaz crystal is mounted, the distance between the source and the topaz crystal varies as much as 4 in., and thus the distance between the collimator and topaz crystal should vary by 4 in. This adjustment was not incorporated into the original design, and thus it is necessary to position the collimator by hand to an approximately correct radial distance for various small portions of the spectrum to obtain precise intensity measurements.

In addition to the divergence effect in the horizontal plane, there is also one in the vertical plane, that is, a variation in the transmission of the instrument for the rays not parallel to the plane of motion of the source. When the source is near the topaz crystal, a larger vertical angle is accepted than when the source is far away. However, if the collimator is positioned properly as defined in the preceding paragraph, it may be shown that this "height effect" almost disappears and for accurate intensity measurements it can be allowed for.

By means of a synchronous motor (Fig. 1) (8), a gear box (9), and a worm drive (10), the source (11), which is mounted on the source arm (12), is made to rotate at a slow uniform rate. By the shifting of levers in the gear box, sweeping rates of 0.3°, 1.5°, 6.0°, and 30.0° Bragg angle per hour can be obtained. In order to have the detector arm (13) make the same angle with the reflecting planes as the source arm (12), the detector arm is linked to the source arm by two steel bands (14) around a pair of drums on the axes of rotation of the respective arms (15). So that the sample will always present the same side to the diffracting crystal, the sample housing is made to rotate in synchronization with the sample arm by the use of another steel band (16). A calibrated scale which reads in arbitrary units of angle plus a vernier (17) on the sample arm permit the reading of the Bragg angle to 0.005°. The scale and vernier (18) by which the detector position is read, are used for aligning the spectrometer.

The pulses from the scintillation counter pass through a preamplifier (19) and a main amplifier before being fed into a pulse height discriminator. Using this differential pulse height analyzer, a small range in pulse amplitude is selected to include the radiation being Bragg reflected. By using a detecting system of this nature, the background is kept down to about 30 counts/min in the 20-kev region (source in the spectrometer, but no peak being Bragg reflected). After discrimination, the pulses are fed into a count-rate meter and a scaler. A Brown chart recorder records the count-rate meter output, while a digital recorder prints the scaler reading at preselected intervals. If the intensity of the line being swept has a counting rate 50 percent higher than the background (defined as the average of the counting rates on the two sides of a peak) or larger, it has usually been found preferable to utilize the chart recorder. This does not reduce the accuracy of the measurements, and it is much less time consuming than taking data from the digital printer. In order to record the angular setting of the spectrometer source arm on the Brown chart at periodic intervals the spectrometer scale is read and the input terminals

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[17] The geometry of the spectrometer is such that the diffracted beam and the undiffracted beam (which is about 100 times as intense as the diffracted beam) would both strike the detector for energies higher than 30 kev unless a collimator is placed over it which will discriminate against all radiation except from a preferred direction.

of the Brown chart are momentarily shorted, causing the pen to drop to zero and return very rapidly, thus providing a sharp reference mark. When the counting rate of a line is too low to obtain good statistics by the continuous sweep method, the spectrometer angle is set by hand to 15 or 20 points spanning a peak, and counts are taken at each point. The digital recorder with the continuous sweep method is used primarily to establish the existence of very weak peaks prior to the hand setting method. It has been found that the worm drive is not uniform, and it is necessary to read the sample arm scale every 5 or 10 minutes for precise measurements.

In order to obtain the maximum resolution from a given sample, it is necessary to have the source perpendicular to the plane of the instrument and to have its geometrical center above the center of rotation of the sample housing. A new sample holder has been designed and constructed by F. Hartford which gives the necessary degrees of freedom. The sample, which is contained in a capillary, is taped to a milled groove cut in a Lucite block which is held in a pair of vertical brass ways by a pair of spring clips. Since the groove and ways are made accurately vertical and the groove is located tangentically over the center of rotation of the sample housing, it is necessary to have only two adjustments, one radically along the sample arm and one vertically. It has been found advisable to make the final precise adjustment by sweeping through a strong single diffraction peak, adjusting the sample position (including rotation of the sample capillary) to obtain high resolution and symmetrical diffraction peaks.

Since this instrument is of the Cauchois approximate-focus type,[18] the diffraction peaks will have a width which will be a function of their angular position and thus their energy, because of a geometrical defocusing due to the deviation of the neutral axis of the bent crystal from the focal circle. This aberration is called the Cauchois width[19] and is defined by

$$d\theta = \sin\theta(1-\cos a)/\cos(a+\theta),$$

where θ is the angle the source arm makes with the crystal perpendicular and a is the half-angle the diffracting crystal intercepts from the source when the source arm is perpendicular to the crystal. This broadening effect will have an effective width on the focal circle on side A of the reflecting planes (see Fig. 2) of zero at 10 kev to 0.011 in. at 200 kev and 0.021 in. on side B of the reflecting planes at 18 kev. The experimental data indicate that this broadening is the major source of aberration in the instrument.

In order to mount sources of moderate specific activity (millicuries per milligram) which can utilize the expected resolving power and for considerations of safe mounting of the intense (frequently alpha-emitting) sources required (∼1 millicurie), the activity is usually sealed in a fine (preferably 10–15 mil i.d.) glass capillary. With sources of very high specific activity (millicuries per microgram), the source is concentrated into a small quantity of solution and then deposited on a fine strip of filter paper. This filter paper is then clamped between two Lucite jaws and placed in the spectrometer so that the thickness of the paper serves as the source width, and thus a dry source only about 0.005 in. thick is easily prepared. It should be pointed out that intense alpha sources sealed in capillaries with water present will decompose the water and build up sufficient pressure to rupture the capillary. For safety considerations, the Lucite jaws that hold the filter paper are inside of a hollow block of Lucite, and after the source is in place, a piece of thin plastic (0.0005-in. mylar) is fastened to the front of the block. It has been found necessary to place a lead slit (20) about one-half inch in front of the source (11) to cut down the scattering from the Lucite jaws. A pair of lead jaws instead of Lucite jaws was constructed to hold the source, but the fluoresence of the Pb was intense enough to give the Pb x-ray lines with an intensity of about 200 counts/min. With a source mounted in this manner, resolutions of 0.25 percent (full width at half-height) have been obtained in the 15-kev region.

The instrument was originally designed to use the (310) planes of quartz, which are perpendicular to the cleavage plane. Since the (303) planes of topaz are at an angle of 29° to the cleavage plane, it is possible to use a set of (303) planes either to the right or left of the crystal perpendicular. In the preliminary calibration work, it was found that the scale to the left of the perpendicular was not linear, and thus this side of the spectrometer was not used subsequently. Rather than try to establish the orientation of the diffracting planes, a "line" is reflected off one side of the (303) planes and then reflected from the other side of the same set of planes, and the Bragg angle is taken as one-half of the angle between the two reflections. Confirmation that the source is located properly (on the axis of rotation of the sample table) is obtained by comparing the (303) orientation obtained from each pair of lines. Individual center lines usually lie within 0.002° of the average of all of the center lines. Owing to the asymmetry of the (303) orientation, energies down to about only 17 kev are measurable on side B (see Fig. 2) of the reflecting planes. For a line of energy lower than 17 kev, its position is measured on side A of the reflecting planes, and then the average of the center lines obtained from all the higher energy lines is taken as the zero for the Bragg angle.

In running a spectrum, each peak is swept a number of times, and it has been found that the angular positions are reproducible to about 0.002° Bragg angle. To obtain the best accuracy, the spectrum is swept at

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[18] Y. Cauchois, Compt. rend. **194**, 1479 (1932); **194**, 362 (1932); **199**, 857 (1943); J. phys. et radium 3, 320 (1932); 4, 61 (1933); Ann. phys. 1, 215 (1934).

[19] J. W. M. DuMond, Rev. Sci. Instr. **18**, 626 (1947).

TABLE I. Reflections from selected topaz planes.

Planes	Bragg angle (1)	Intensity Bragg peak (2)	 Diffuse peak (3)
101	1.5°		120
202	2.9°		180
303	4.4°	5000	30
404	5.9°	50	40
505	7.4°		60
606	8.9°	200	...
to			
13013	19.5°	<5	<5
100	1.3°		100
200	2.5°		200
300	3.8°		60
400	5.1°	600	10
500	6.5°		30
600	7.7°	400	5
700	9.0°		30
800	10.3°		...
900	11.6°		5
1000	12.9°	130	...
to			
3000	42.3°	<5	<5

the slowest sweep speed, 0.3° per hour, and thus it takes about 150 hours to make one complete sweep of the spectrum on both sides of the diffracting planes. By using pulse-height discrimination as described above, the background varied from about 70 counts/min at 60 kev (using 15 baffles in the collimator) to about 30 counts/min at 15 kev (using 7 baffles in the collimator) for the Am^{241} source used in these experiments. (This background range depends strongly on the spectrum of the sample.) Background is defined as the average of the intensity on the two sides of a diffraction peak.

The lower energy limit of the instrument is primarily due to the absorption in the topaz crystal, which absorbs 90 percent of the radiation at 8 kev, at which energy the absorption in the air and the self-absorption in the sample also becomes quite large. The upper energy limit is determined by the ability of the collimator to discriminate between the direct and the diffracted beam.[17] The highest-energy line thus far measured has been 120 kev, but from purely geometrical considerations, the upper limit should be about 200 kev. One means of extending the upper limit of the instrument would be to charge the geometry of the collimator by increasing the number of baffles and/or lengthening it. An alternative means would be to use reflections from planes other than the (303) planes with higher dispersion. Table I shows the results of an investigation of the reflectivity of the ($h0l$) and the ($h00$) planes with the 60-kev gamma ray of Am^{241}. Column (1) lists the angle at which the 60-kev diffraction peak was observed. Two distinct types of diffraction maxima were observed, the usual Bragg peak which had a full width at half-height of 5.4 minutes (column 2), and a superimposed diffuse maximum with a full width at half-height of 38 minutes (column 3). After removing all possible sources of scattering in the instrument, the intensity and shapes of the peaks were unaltered. The centers of the diffuse maxima lie just where a Bragg reflection would be expected for the various order reflections. It is believed that these diffuse maxima are due to the thermal waves that exist in the crystal lattice.[20] From Table I it may be seen that by using the (1000) planes with a 40-fold loss in intensity, the energy range of the spectrometer could be extended to about 3 times its present limit or 600 kev.

CALIBRATION

In order to establish the reliability and self-consistency of the instrument, a number of known x-ray lines have been measured. The first calibration lines used were the $K\alpha_1$ and $K\beta_1$ lines of zirconium. A 0.010-in. wire of zirconium metal, suspended from its ends in the source holder, was irradiated from above by a tungsten target x-ray tube operating at 25 kev and 300 μa, which readily excited the K-radiation of zirconium. The energies of the two x-ray lines due to fluorescence were measured and found to disagree with the Cauchois and Hulubei[21] values. It was found necessary to change the accepted value of the topaz spacing from 1.356 A[22] to 1.3525 A. Although this new choice of lattice constant was made with the measurement of only two lines, it has also given the correct energy values in subsequent calibrations on other elements (Th and La). The final adjusted values obtained for the $K\alpha_1$ and $K\beta_1$ lines of zirconium are 15.772±0.003 kev and 17.671±0.003 kev, respectively, in comparison to the Cauchois and Hulubei[21] values of 15.775 and 17.668 kev, using $E\lambda_s = 12372.2$ kev x-units to convert from Siegbahn units to kev as given by DuMond and Cohen.[23] The most recent value for the topaz (303) spacing has been determined by Silfverberg[24] on a double-crystal spectrometer to be 1.352902 A, agreeing to 0.0004 A with the value used in this work.

A sample of thorium in the form of ThO_2 was loaded into a capillary with an internal diameter of 0.012 in. and irradiated with the x-ray tube as described above. The measured values of the $L\alpha_1$ and $L\beta_1$ lines of thorium are 12.964±0.005 kev and 16.207±0.003 kev, respectively; the Cauchois and Hulubei[21] values are 12.968 kev and 16.202 kev.

Since a power supply with sufficient voltage to excite the lanthanum K x-rays was not available, a sample of La_2O_3 in a capillary was made to fluoresce with the 60-kev gamma radiation from a sample of Am^{241} in an adjacent capillary. The measured values of

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[20] R. W. James, *The Optical Principles of the Diffraction of X-Rays* (G. Bell and Sons, London, 1948), pp. 239–64.

[21] Y. Cauchois and H. Hulubei, *Constantes Selectionnees, Longueurs D'Onde Des Emissions X Et des Discontinuites D'Absorption X* (Hermann and Company, Paris, 1947).

[22] Hermann, Lohrmann, and Philipp, Strukturbericht **2**, 516 (1928–1932).

[23] J. W. M. DuMond and E. R. Cohen, Revs. Modern Phys. **25**, 691 (1953).

[24] L. Silfverberg, Arkiv Mat. Astron. Fysik **B31**, No. 10 (1945).

the $K\alpha_1$ and $K\alpha_2$ energies are 33.440 ± 0.008 kev and 33.031 ± 0.015 kev, respectively; the Cauchois and Hulubei[21] values are 33.442 kev and 33.035 kev.

Since the second-order reflection of the sixty-kev transition (8.9° Bragg angle) of Am^{241} was found to agree with the first-order reflection (4.4° Bragg angle), to within the experimental errors, and calibrations at 33 kev (7.9° Bragg angle), at 16 kev (16.4° Bragg angle), and at 13 kev (20.7° Bragg angle) are in excellent agreement with the Cauchois and Hulubei[21] table values, it is assumed that the entire scale of the spectrometer is linear. It should be noted that the experimental error in the energy varies as

$$dE/E = d\theta/\tan\theta.$$

Thus with a constant angular error of 0.002°, the error in the energy at 30 kev will be 7 ev, while the error at 60 kev will be 28 ev. Thus whenever the second-order reflection is strong enough to obtain good statistics, the energy accuracy can be improved.

Am^{241} SPECTRUM

The sample consisted of 1.5 milligrams of finely powdered AmO_2 loaded into a quartz capillary with an internal diameter of 0.33 mm and a wall thickness of 0.06 mm. The sample filled a 1.7-cm length of the capillary; the calculated bulk density was taken as unity. In the preparation of the sample, specific steps were taken to remove any rare earths, since the K x-rays of these elements lie between 30 and 40 kev, which has proved to be an interesting region in the Am^{241} spectrum (see Introduction). A spectrographic analysis showed that the only rare earth present was La, with a concentration of 0.1 percent. The K-radiation emitted from this quantity of La, which would be excited by the strong 60-kev gamma of the Am^{241} was calculated to be equal to less than one part in ten thousand of the 60-kev radiation. (A 33-kev gamma was found in the Am^{241} spectrum with an intensity of one part in two hundred of the 60-kev radiation.)

With a sample of this diameter, the resolution is still mainly a function of the source width (resolution being defined as the full width of the line at half-maximum). Resolutions of about 0.5 percent and 1.8 percent were obtained in the 17-kev and 60-kev regions, respectively. In other experiments, with sources of higher specific activity, where the source is deposited on a piece of filter paper, the resolution in the 17-kev region has been improved to 0.25 percent. Figure 3 is a picture of a chart record taken with the spectrometer sweeping at 20 times the normal sweep rate and with the pulse height discriminator set to accept all pulses between 10 and 60 kev. From the figure it appears that it would be difficult to establish the existence of any weak gamma rays in the 30- to 50-kev region, but as previously stated (Apparatus), when the pulse height

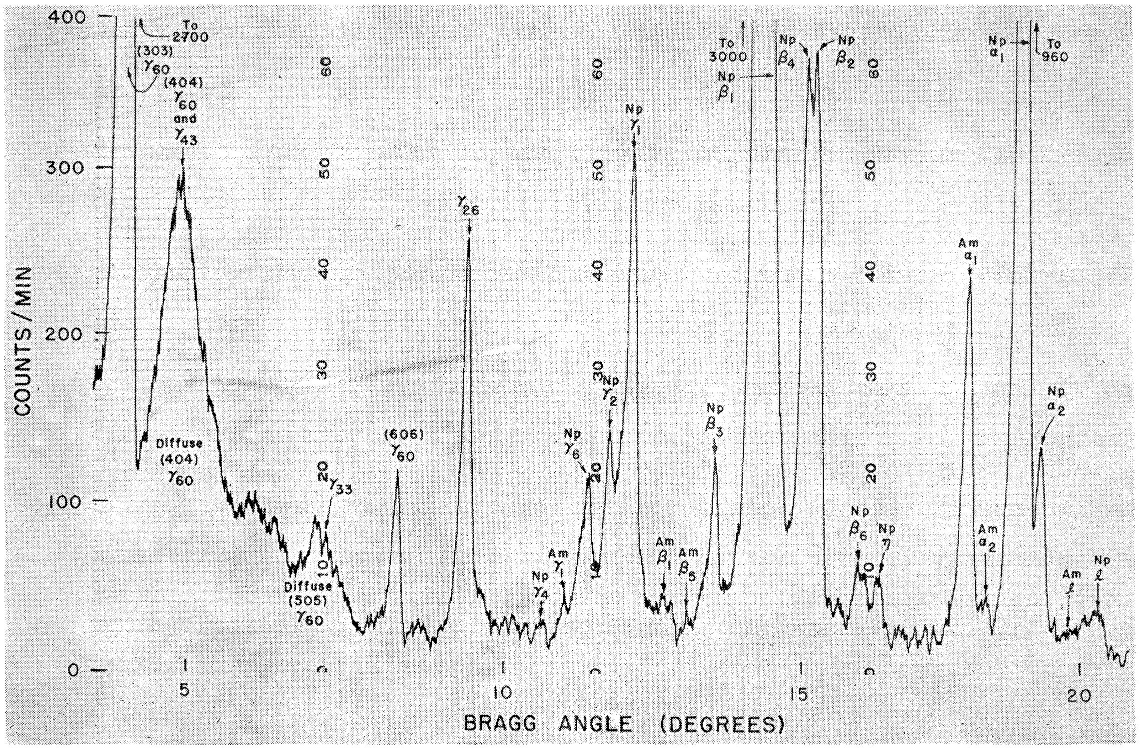

FIG. 3. Chart record of the complete Am^{241} spectrum (pulse height selection not used). Only the more prominent lines have been labeled since the weak lines are indistinguishable from the statistical fluctuations in the background in this plot.

TABLE II. The Am^{241} gamma rays observed on the bent-crystal spectrometer.

Energy (kev)	Background[a] (counts/min)	Observed intensity[b] (counts/min)	Correction[c] factor	Corrected[d] intensity
26.363±0.014	40	520	1.55	8.2
33.199±0.021	39	39	1.32	0.5
43.463±0.085	65	30	2.17	0.6
59.568±0.017	70	5220	1.88	100

[a] Background computed by averaging counting rates on both sides of each peak.
[b] Background subtracted.
[c] Attenuation corrections as explained in text.
[d] Relative to the 60-kev line.

discriminator is set so that any radiation which is being Bragg reflected in orders other than the one under study is rejected, the background in the 30 to 50-kev region is lowered to about 20 counts/min.

Table II lists the energies and the relative intensities of the gamma rays that were observed on the bent-crystal spectrometer. The correction factor (column 3) is made up of the following attenuation factors: self-absorption in the sample, absorption in the capillary wall, absorption in the air, absorption in the topaz crystal, reflectivity of the topaz as a function of energy, absorption in the detector window, and efficiency of the detector. Of these the least certain are the self-absorption and the reflectivity corrections. Table III lists the corrections that were applied to the 17.760-kev $L\beta_1$ x-ray of Np and the 59.568-kev gamma ray.

Local density variations from the average value limit the accuracy of the correction for the self-absorption. In addition, the mass absorption coefficients for the transuranium elements have not been measured, and thus it is necessary to derive them by extrapolations from thorium and uranium. A third difficulty is that the L x-rays of Np lie within the L-absorption edges of Am, which further limits the reliability of the corrections. In consideration of the above difficulties, it is estimated that the self-absorption corrections are reliable to only about 20 percent.

At the present time the reflectivity of the topaz is questionable. Lind *et al.*[25] have measured the reflectivity of the (310) planes of an elastically bent quartz crystal and found it to obey a $1/E^2$ dependence (over the range from 25 kev to 1.3 Mev), whereas an unbent quartz crystal obeys a $1/E$ dependence. In order to roughly check the reflectivity of the (303) planes of topaz, a pulse height spectrum of the bent-crystal spectrometer sample was taken with the same scintillation detector as used in the bent-crystal spectrometer, and the observed intensities were compared to those from the diffracted beam in the bent-crystal spectrometer. In order to cut down the counting rate to a reasonable value, the pulse height curve was taken with the sample several feet from the detector. The same absorption corrections apply to this pulse height experiment as to the bent-crystal spectrometer data except for the air absorption and the topaz crystal absorption corrections. The absorption coefficients of the topaz have been measured at a number of monochromatic energies (13, 17, and 60 kev) in the spectrometer and agree to within 5 percent of the values computed from the known chemical composition $\{[Al(FOH)]_2SiO_4\}$ and the Victoreen[26] absorption tables. The correction for the air absorption in the "pulse height" experiment is considerably more difficult. The pulse height spectrum (Fig. 4) consists of essentially two peaks, the 60-kev peak and a broad peak centering around 18 kev consisting of the L x-rays and a hump on the high-energy side due to the 26-kev gamma and the 60-kev escape peak. To simplify the calculation of the air absorption corrections for each of the components of the low-energy peak, it was assumed to consist of the three prominent components observed in the bent-crystal spectrometer (13, 17, and 26 kev), and the absorption corrections were made for these components. The intensity of the escape peak was taken from a pulse height spectrum of the 60-kev gamma diffracted beam and found to be 12.7 percent, which agrees perfectly with Axel's[27] computed value for good geometry (radiation impinging perpendicular to the crystal face). Applying these corrections, the ratio of the area of the high-energy to the low-energy peak is 1.7. The sum of the intensities of all the observed lines in the bent-crystal spectrometer which contributed to the low-energy peak in the pulse height spectrum was compared with the intensity of the 60-kev (plus the 43-kev gamma which was not resolved from the 60-kev line in the pulse height spectrum). Assuming a $1/E^2$ dependence for the reflectivity, a ratio of 4.2 was obtained, while a $1/E$ dependence gave a value of 1.4. It thus seems evident that the reflectivity of the (303)

TABLE III. Example of the various intensity corrections that are used.

Line	Energy (kev)	Self-absorption	Capillary wall	Air	Topaz absorption	$1/E$ reflectivity	Collimator[a]	Detector efficiency	Total correction
$L\beta_1$	17.760	1.4	1.04	1.08	1.32	0.30	1.00	1.03	0.64
γ_{60}	59.568	1.1	1.00	1.01	1.01	1.00	1.66	1.01	1.88

[a] A function of the number of baffles used.

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[25] Lind, West, and DuMond, Phys. Rev. **77**, 475 (1950).

[26] J. A. Victoreen, J. Appl. Phys. **20**, 1141 (1949).

[27] P. Axel, Brookhaven National Laboratory Report BNL-271, September, 1953 (unpublished).

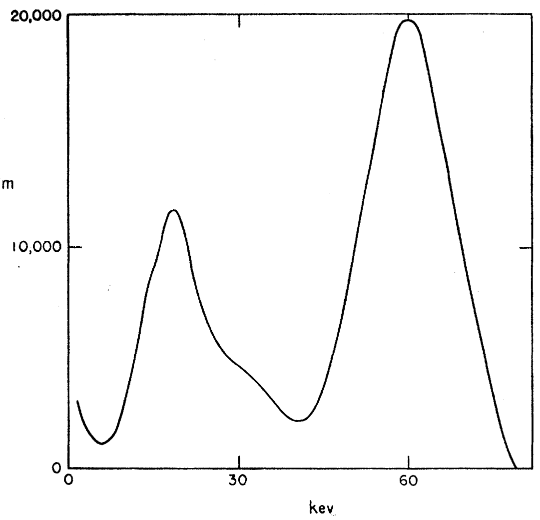

FIG. 4. Pulse height spectrum of the Am^{241} sample used in the bent-crystal spectrometer.

planes of topaz exhibits an approximately $1/E$ dependence rather than $1/E^2$. It is intended to measure this reflectivity dependence more precisely in the near future, but for the present, the $1/E$ dependence will be used.

The gamma-rays listed in Table II fit well into the Np^{237} level scheme proposed by Wagner *et al.*[15] and Asaro *et al.*[2] as shown in Fig. 5. The 33.199-kev gamma ray has not been previously observed. It is to be noted that its intensity of 0.5 percent of the 60-kev gamma is somewhat higher than the upper limit of 0.1 percent set by Newton and Rose[12] on a proportional counter, It is likely that this is a true Am^{241} gamma ray rather than the lanthanum K x-rays, since, as already noted (see Calibration), the $K\alpha$ doublet of La was resolvable into its components, whereas the 33-kev peak in the Am^{241} spectrum is very clearly a single line with the expected width for this energy region. Also as noted above, the maximum La fluorescence from this sample would only be 0.01 percent of the 60-kev gamma intensity. It should also be observed that the sum of the 33-kev and 26-kev gamma-ray energies agrees to within 6 ev with the 60-kev gamma energy.

The 43-kev gamma ray listed in Table II has not previously been observed, but conversion lines due to this energy transition have been observed by Passell[13] and Wagner *et al.*[15] Considerable difficulty was encountered in measuring this line. The diffraction maximum was a composite of two nearly superimposed peaks. It was found that by adjusting the pulse height selector so that practically no 60-kev pulses were passed, the high-energy component disappeared and the low-energy peak was clearly a single one with

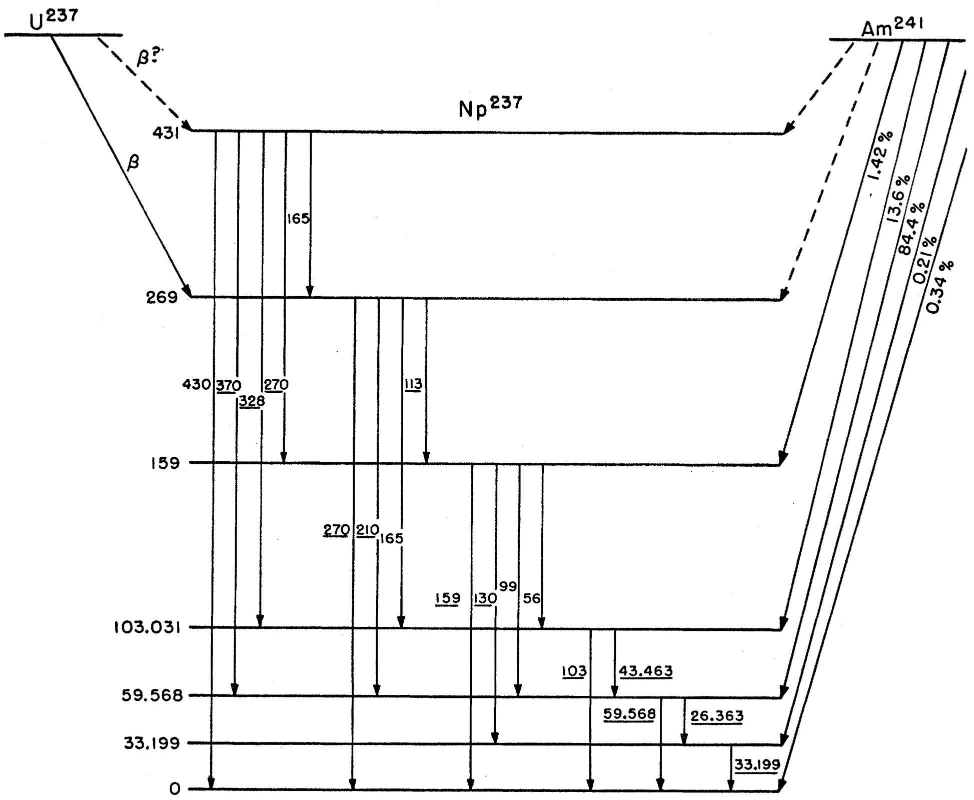

FIG. 5. Energy level scheme of Np^{237} (gamma transitions underlined were observed in this work).

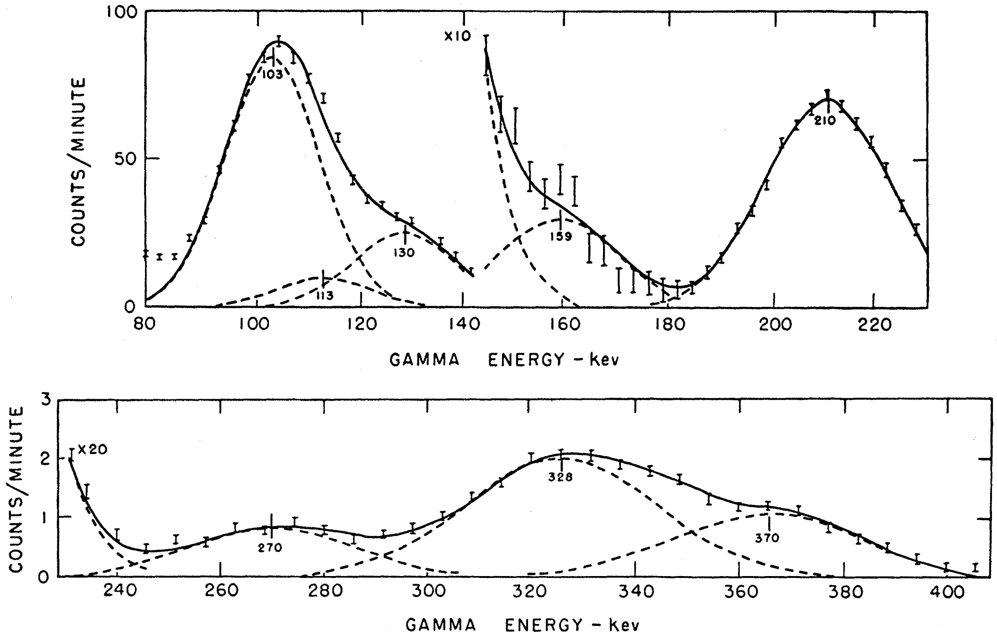

FIG. 6. Pulse height spectrum of Am^{241} taken on the 20-channel scintillation spectrometer through 1.63 g/cm^2 of Ag.

an energy of 43.463 kev. By setting the pulse height selector so that all the 43-kev pulses were eliminated, the low-energy peak disappeared. The high-energy peak was identified as the (404) reflection of the 60-kev gamma ray.

Gamma rays of 56 kev and 70 kev are possible transitions in the proposed level scheme, but upper intensity limits of about 10^{-4} gamma ray per disintegration may be set for these regions with the bent-crystal spectrometer. Since the transmission of the bent-crystal spectrometer is not high enough to detect the existence of the higher-energy gamma rays reported by Passell[13] and Jaffe,[14] a pulse height spectrum of the sample was taken with a sodium iodide scintillation spectrometer and 20-channel analyzer. The energy scale and linearity of the scintillation counter were established by taking pulse height curves of the 59.6-kev line of Am^{241},[28] the 87-kev line Cd^{109},[29] the 104-kev line of Gd^{153},[30] the 134-kev line of Ce^{144},[31] and the 138-kev, 269-kev, and 405-kev lines of Se^{75}.[32] To prevent the intense low-energy radiation from overloading the detecting system, a 1.63-g/cm^2 silver absorber was placed over the detector. Figure 6 shows the pulse height spectrum obtained from a 20-microgram sample of Am^{241} using a one-half inch thick sodium iodide crystal at about 1 percent geometry. The pulse height curve was unfolded into the component parts indicated by using the Wagner *et al.*[15] data as a guide, assuming that the resolution of the scintillation detector follows a $1/E^2$ dependence, and a single line has a Gaussian shape. Table IV lists the energies and intensities (if one assumes 0.40 60-kev gamma per disintegration)[9] of the gamma rays observed with Jaffe's[14] values listed in columns 2 and 4. The 113, 270, 328, and 370-kev gamma rays have not been previously observed in the Am^{241} decay, but conversion lines for these gamma-ray energies have been reported in the U^{237} spectrum by Wagner *et al.*[15] with the exception of the 113-kev gamma. Figure 5 depicts the energy level scheme of Np^{237} using all the available data. It is still quite tenuous since some of the gamma rays could be transitions between more than one pair of levels and the

TABLE IV. The Am^{241} gamma rays observed on the 20-channel scintillation spectrometer.

Energy (kev) Observed here	Jaffe[a]	Intensity (per/disintegration) Observed here	Jaffe[a]
103±2	102	4.0×10^{-4}	5.6×10^{-4}
113±5		3.3×10^{-5}	
130±4	128	5.3×10^{-5}	5.6×10^{-5}
159±10	168	4.4×10^{-6}	6.4×10^{-6}
210±5	207	8.4×10^{-6}	7.6×10^{-6}
270±10		1.1×10^{-6}	
328±10		3.0×10^{-6}	
370±10		1.7×10^{-6}	

[a] See reference 14.

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[28] See Table II.

[29] Huber, Humbel, Schneiden, and deShalit, Helv, Phys. Acta **25**, 3 (1952).

[30] Cork, LeBlanc, Nester, and Stumpf, Phys. Rev. **88**, 685 (1952).

[31] F. T. Porter and C. S. Cook, Phys. Rev. **87**, 464 (1952).

[32] Jensen, Laslett, Martin, Hughes, and Pratt, Iowa State College Report ISC-252 (unpublished).

higher energy levels are not known very accurately. In attempting to compute reasonable values for the conversion coefficients, it turns out that the intensity of the source and the absolute geometry of the spectrometer must be known to a high degree of accuracy (1 percent). Since this data is not readily obtainable, the conversion coefficients will be tabulated by Freedman *et al.*[33] using their conversion electron data and the relative gamma intensities reported here.

Table V lists the L x-ray lines found due to internal conversion in Np^{237} together with their intensities. The measured energies average about 19 electron volts higher than the extrapolated values in the table of Hill *et al.*[34] Since the latter energy values are not claimed to be reliable to 20 ev, this 19-ev discrepancy is not deemed significant. Upon summing the intensities of the x-rays arising from the three L levels (Table VI), the ratios of the radiative transitions from the three levels are obtained:

$$L_1:L_2:L_3=1.0:4.6:3.6.$$

To determine the relative abundances of initial L_1, L_2, and L_3 vacancies, it is necessary to allow for the Coster-Kronig and Auger transitions. Kinsey[35] has calculated the fluorescence yields from the ratios of the radiation widths to the total widths of the levels, obtaining 0.16, 0.59, and 0.41 for the fluorescence yields of the L_1, L_2, and L_3 levels, respectively, in uranium. Since these values vary only slightly with atomic number, they are applied to neptunium. He also assumes that the Coster-Kronig transitions from the L_1 to L_2 and L_2 to L_3 are negligible compared to the L_1 to L_3 transition. Since the Coster-Kronig effect increases the level width from which it originates, Kinsey computed the probability of this transition by comparing the radiation width of the L_1 level in tantalum, where the effect is small, to the radiation width of the L_1 level in uranium and obtained a value of 0.60. Kinsey expresses the following equations:

$$q_1=u_1f, \tag{1}$$

$$q_2=u_2f_2, \tag{2}$$

$$q_3=f_3(A_{13}u_1+u_3), \tag{3}$$

where q_1, q_2, and q_3 are the number of quanta observed, f_1, f_2, and f_3 are the fluorescent yields, and u_1, u_2, and u_3 are the number of initial vacancies in the L_1, L_2, and L_3 shells, respectively. A_{13} is the probability of the L_1-L_3 Coster-Kronig transition. Using Kinsey's values for A_{13}, f_1, f_2, and f_3, the ratios of initial L vacancies are:

$$L_1:L_2:L_3=1.0:1.3:0.8.$$

TABLE V. The Np L x-rays observed on the bent-crystal spectrometer due to internal conversion.

Line	Experimental energy (kev)	Extrapolated energy[a] (kev)	Background (counts/min)	Observed intensity (counts/min)	Correction factor	Corrected relative intensity[b]
l	11.889±0.006	11.87	40	60	2.40	1.3
α_2	13.776±0.003	13.76	60	285	1.09	2.8
α_1	13.961±0.003	13.95	60	2760	1.01	25.2
η	15.876±0.003	15.85	80	150	0.83	1.2
β_6	16.139±0.024	16.11	90	80	0.75	0.5
β_2	16.857±0.006	16.84	130	1020	0.70	6.5
β_4	17.075±0.006	17.05	140	681	0.68	4.2
β_5	17.516±0.009	17.51	120	180	0.62	1.0
β_1	17.764±0.003	17.74	120	6300	0.64	36.4
β_3	18.001±0.005	17.98	200	580	0.63	3.3
γ_5	20.124±0.040	20.09	95	35	0.88	0.3
γ_1	20.796±0.005	20.77	120	1020	0.92	8.5
γ_2	21.108±0.020	21.09	140	110	0.94	0.9
γ_3	21.340±0.017	21.33	120	80	1.47	1.1
γ_6	21.476±0.015	21.49	50	170	1.47	2.2
γ_4	22.200±0.022	22.16	70	80	1.39	1.0

[a] See reference 34, table values.
[b] Relative to 60-kev line =100.

Some insight as to the correctness of Kinsey's results may be obtained from the relative intensity of the Np $L\beta_2$ satellite[36] that was observed at 86±23 volts higher than the parent line. A satellite of this energy is ascribable to a Coster-Kronig transition as follows: An L_1 vacancy is created due to internal conversion of a gamma-ray, and an L_3 electron falls into the L_1 level, the difference in energy being used to eject an M electron. With the atom in this doubly ionized state, an N electron falls into the L_3 vacancy. The energy of this transition will be emitted as an x-ray of higher energy than if the M vacancy were not present. The energy difference between the normal x-ray line (following an initial L_3 vacancy) and its satellite is

TABLE VI. Designation of the L x-ray line transitions and their relative intensities in Am and Np.

Line	Transition	Intensity[a] Np	Am
β_4	L_1-M_2	4.2	
β_3	L_1-M_3	3.3	0.4
γ_2	L_1-N_2	0.9	0.3
γ_3	L_1-N_3	1.1	
γ_4	L_1-O_2	1.0	
η	L_2-M_1	1.2	
β_1	L_2-M_4	36.4	2.5
γ_5	L_2-N_1	0.3	
γ_1	L_2-N_4	8.5	1.0
γ_6	$L_2-O_{4,5}$	2.2	
l	L_3-M_1	1.3	0.2
α_2	L_3-M_4	2.8	0.8
α_1	L_3-M_5	25.2	6.4
β_6	L_3-N_1	0.5	
β_2	L_3-N_5	6.5	
β_5	$L_3-O_{4,5}$	1.0	0.4

[a] Relative to 60-kev line =100.

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[33] Freedman, Porter, and Wagner, to be published in Physical Review.

[34] Hill, Church, and Mihelich, Rev. Sci. Instr. **23**, 523 (1952). Even though the instrument was calibrated by using the Cauchois and Hulubei[21] values, it seems correct to compare the Np x-ray energy values with the Hill *et al.* values since these values are about one or two ev higher than the Cauchois values for Zr and Th and 10 to 15 ev lower for the La.

[35] B. B. Kinsey, Can. J. Research **A26**, 404 (1948).

[36] There is also evidence that there is a satellite on the high-energy side of the $L\beta_5$ line, but since it is close to the intense $L\beta_1$ line, it is not possible to obtain any significant information except that it exists. It would be impossible with the present spectrometer and source to see any $L\alpha$-satellites if they existed, because the energy separation would be less than 10 ev.

TABLE VII. The Am L x-rays observed on the bent-crystal spectrometer due to fluorescence by the gamma rays.

Line	Experimental energy (kev)	Extra-polated energy (kev)	Back-ground (counts/min)	Observed intensity (counts/min)	Correc-tion factor	Corrected relative intensity[a]
l	12.383 ±0.007	12.38	40	15	1.53	0.2
α_2	14.416 ±0.030	14.41	60	90	0.96	0.8
α_1	14.629 ±0.003	14.62	60	760	0.93	6.4
β_5	18.408 ±0.008	18.40	110	65	0.59	0.4
β_1	18.871 ±0.005	18.83	95	300	0.90	2.5
β_3	19.105 ±0.008	19.10	95	40	0.91	0.4
γ_1	22.076 ±0.018	22.04	24	80	1.40	1.0
γ_2	22.359 ±0.025	22.35	80	20	1.43	0.3

[a] Relative to 60-kev line = 100.

given by:

$$dE=[(E_{Mx})_{Z+1}-(E_{Mx})_Z]-[(E_{Ny})_{Z+1}-(E_{Ny})_Z],$$

where $(E_{Mx})_{Z+1}$ is the binding energy of an electron in the M_x subshell of a $Z+1$ atom, and the other symbols have a corresponding meaning. The equation is written on the assumption that when an electron is missing in an inner shell, an electron in an outer shell has the energy of an electron in the corresponding shell of a $(Z+1)$ atom. The energy separations of the satellites due to the absence of an M_3, M_4, or M_5 electron are 89, 89, or 79 ev, respectively, and thus it may be concluded that the prominent component of the satellite was due to one of these vacancies. If it is assumed that the transition probabilities for the various lines do not change when the atom is in the doubly ionized state, then one may use the intensity of the $L\beta_2$ satellite as a measure of the Coster-Kronig transition probability. Using Eq. (3), q_3 (number of observed quanta from L_3) will be equal to the sum of the intensities of $L\beta_2$ and its satellite, and $A_{13}u_1$ (the number of L_3 lines arising from Coster-Kronig transitions from L_1) will be the intensity of the satellite; the data determine a ratio:

$$q_3/A_{13}u_1=3.27.$$

Since q_3 is known and u_1 has been computed from the experimental data, and using Kinsey's value for the L_1 fluorescent yield, one obtains a value of 0.17 for the Coster-Kronig transition probability A_{13}, where Kinsey obtained a value of 0.60. This experimental result is partially substantiated by Jaffe,[14] who set an upper limit of 0.40 on this coefficient in neptunium. If the transition probabilities of the lines change when the atom is in the doubly ionized state, and specifically if the probability for the $L\beta_2$ transition decreases with respect to the other transition probabilities, then the derived value for the Coster-Kronig coefficient would be altered in the direction to agree with Kinsey's calculations. Using the derived value ($A_{13}=0.17$), the relative excitations of the three levels become:

$$L_1:L_2:L_3=1.0:1.3:1.2.$$

TABLE VIII. Higher-order reflections in the Am^{241} spectrum. Columns (3) and (4) give the ratio of the (303) reflected intensity to the (606) and (404) reflected intensities, respectively.

Line	Energy (kev)	303/606	303/404
$L\beta_1$	17.760	32	
$L\gamma_1$	20.786	25	
γ_{26}	26.356	21	
γ_{60}	59.563	27	104

Table VII lists the energies and the relative intensities of the americium L x-rays observed due to fluorescence from the gamma rays. The measured energies average about 15 ev higher than the values in the Hill *et al.* table,[34] and thus the Mosley extrapolations are verified within about 15 ev.

Four lines in the Am^{241} spectrum were intense enough to be observed in higher-order reflections than the first (Table VIII). The variation in the ratios is not deemed significant since the higher order reflections were quite weak and were not all clearly resolved from other features of the spectrum. The most reliable ratio is the one obtained for the 60-kev gamma ray.

The author wishes to express his indebtness and thanks to Dr. M. S. Freedman for his untiring assistance and advice throughout the work. The author also wishes to thank Mr. J. Malm for the aid in preparing the sample, Dr. D. W. Engelkemeir for his advice on the scintillation detector problems which have arisen, and Dr. S. Siegel for his helpful discussions on the physics of x-ray spectrometry.

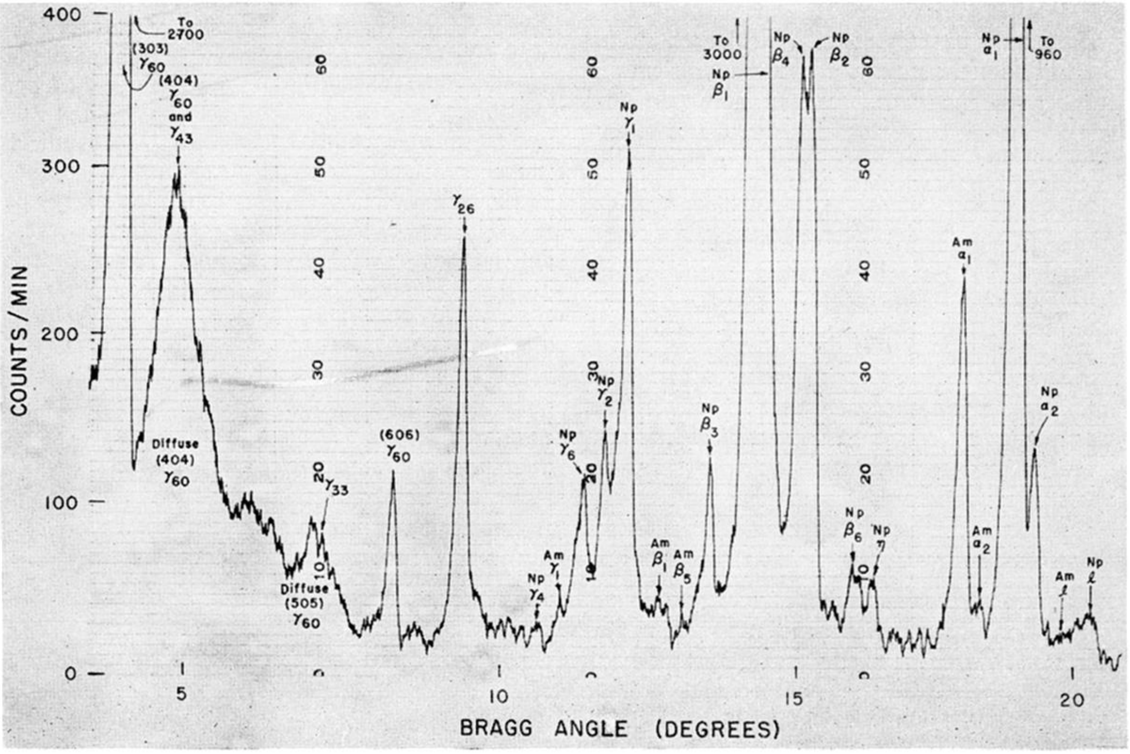

FIG. 3. Chart record of the complete Am^{241} spectrum (pulse height selection not used). Only the more prominent lines have been labeled since the weak lines are indistinguishable from the statistical fluctuations in the background in this plot.