

## The Internal Conversion Electrons of Several Short-Lived Neutron Induced Radioactivities\*†

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Accurate values of the transition energy were measured for seven neutron-induced radioactivities using a permanent magnet beta-ray spectrograph. A special vacuum gate permitted the introduction of short-lived samples within twelve seconds after the end of bombardment without disturbing the vacuum in the spectrograph. Transition energies, accurate to about one percent, were determined as follows: cesium (3 hr.) 128.0 kev, cobalt (10.7 min.) 58.9 kev, columbium (6.6 min.) 41.5 kev, dysprosium (1.3 min.) 109.0 kev, dysprosium (2.6 hr.) 87.8 kev, iridium (1.5 min.) 57.4 kev, thulium (120 day) 84.8 kev. For the five isomeric transitions, approximate values of the relative intensities of the electron lines were determined, and an assignment was made of the multipole order of the radiation, assuming electric multipole radiation.

### 1. INTRODUCTION AND METHOD

TO study the internal conversion electrons of several short-lived radioactivities, a permanent magnet spectrograph, similar to that described by Hill,<sup>1</sup> was constructed. Because of the integrating action of the photographic film detector, short half-life period radioactivities could be studied by using successive radioactive samples to give an accumulated exposure on a single film. A special vacuum gate was used to introduce into the spectrograph a radioactive sample within twelve seconds after the end of its bombardment without seriously altering the vacuum in the spectrograph. The design of this gate is shown in Fig. 1. It consists of a small forechamber separated from the main spectrograph vacuum chamber by a hand operated

gate. A Welch forepump was used to produce the required vacuum of  $10^{-3}$  mm Hg in the main chamber. A second forepump was connected to the forechamber with a hand valve in the pumping line. A circular opening in the forechamber allowed insertion of a sliding cylindrical rod with removable Lucite source holder on its end. An O-ring gasket formed a vacuum seal between the rod and forechamber walls permitting both translational and rotational motion of the rod holding the source.<sup>2</sup> With this arrangement it was possible to insert the rod with the source on it through the O-ring seal into the forechamber and permit rough pumping of the forechamber prior to opening the gate and sliding the rod forward to position the sample in the spectrograph.

The source in powder form was attached to a narrow

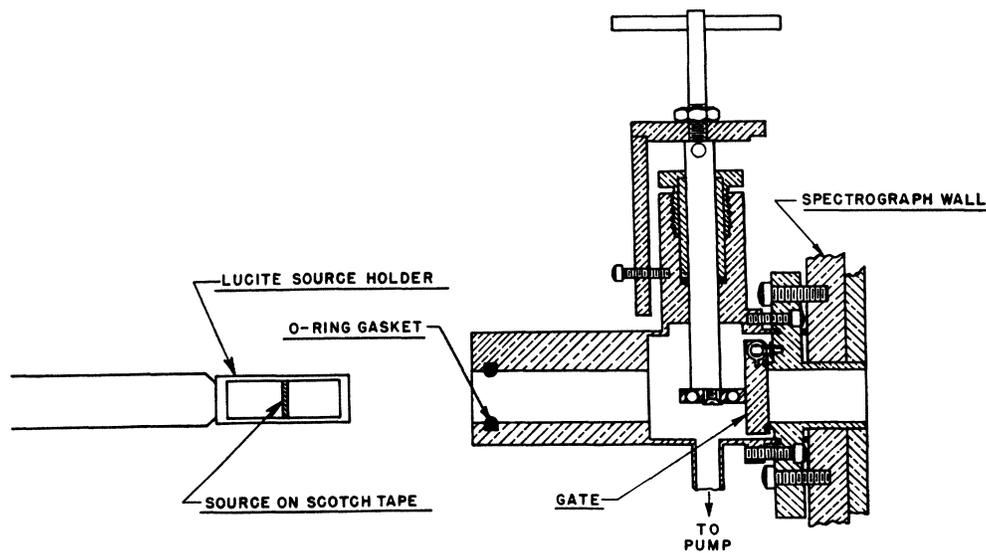


FIG. 1. Vacuum gate for quick source change. This vacuum gate was used to introduce into the spectrograph a radioactive sample within twelve seconds after the end of its bombardment without seriously altering the vacuum in the spectrograph.

\* Extract from a thesis submitted in partial fulfillment of the requirements for the degree of Doctor of Philosophy at the University of Missouri. A preliminary report was given by Caldwell, der Mateosian, and Goldhaber, *Phys. Rev.* **76**, 187(A) (1949).

† This work was carried out at the Argonne National Laboratory under auspices of the AEC.

<sup>1</sup> R. D. Hill, *Phys. Rev.* **74**, 78 (1946).

<sup>2</sup> F. N. D. Kurie, *Rev. Sci. Inst.* **19**, 485 (1948).

TABLE I. Electron and transition energies.

Element	Half-life	$H\rho$ (gauss-cm)	Electron energy (kev)	Conversion shell	Transition energy (kev)
Cs	3 hr.	1067.5	92.0	<i>K</i>	128.0
		1251.1	123.0	<i>L</i>	128.0
		1273.5	127.0	<i>M</i>	128.0
					Av. = 128.0
Co	10.7 min.	781.9	51.1	<i>K</i>	58.8
		835.8	58.2	<i>L</i>	59.0
		839.9	58.7	<i>M</i>	58.8
					Av. = 58.9
Cb	6.6 min.	510.1	22.5	<i>K</i>	41.5
		678.2	39.0	<i>L</i>	41.5
		695.1	40.8	<i>M</i>	41.2
					Av. = 41.4
Dy	1.3 min.	812.0	55.1	<i>K</i>	109.0
		1117.5	100.2	<i>L<sub>I</sub></i>	109.3
		1122.9	101.1	<i>L<sub>III</sub></i>	108.9
		1158.6	107.1	<i>M</i>	108.8
		1167.9	108.7	<i>N</i>	109.0
					Av. = 109.0
Dy	2.6 hr.	616.2	32.5	<i>K</i>	88.2
		982.1	78.8	<i>L<sub>I</sub></i>	88.2
		988.5	80.0	<i>L<sub>III</sub></i>	88.1
		1029.8	85.8	<i>M</i>	87.2
		1039.8	87.2	<i>N</i>	87.4
					Av. = 87.8
Ir	1.5 min.	1734.3	219.1	<i>K</i> (?)	
		722.9	44.1	<i>L<sub>I</sub></i>	57.5
		739.1	46.0	<i>L<sub>III</sub></i>	57.2
		806.6	54.4	<i>M</i>	57.3
		823.0	56.5	<i>N</i>	57.3
					Av. = 57.4
Tm	120 days	515.8	22.9	<i>K</i>	84.4
		954.3	74.8	<i>L<sub>I</sub></i>	85.3
		961.5	75.9	<i>L<sub>III</sub></i>	84.9
		1007.7	82.7	<i>M</i>	84.8
		1018.3	84.3	<i>N</i>	84.8
					Av. = 84.8

piece of Scotch Tape on a source holder which was bombarded with thermal neutrons from the heavy water moderated chain-reacting pile of the Argonne National Laboratory. Since several source holders were used interchangeably, a special jig was constructed for cutting the Scotch Tape to which the source was attached. This jig assured that all sources were the same width and were aligned in the same position with respect to the slit opening of the spectrograph.

The photographic film used was Eastman No-Screen X-Ray film. In order to make approximate measurements of the relative intensities of the various electron lines the film was calibrated by using a thulium source (120-day half-life period). The data of Saxon<sup>3</sup> were used as a "standard" to give the number of electrons at any particular energy.

<sup>3</sup> D. Saxon, and J. Richards, Phys. Rev. **76**, 186 (1949).

Calibration of the magnetic field strength of the spectrograph magnet was made by two independent methods. First, the x-ray critical absorption energies were used to obtain an accurate evaluation of the magnetic field strength in the following way. A provisional assignment of the conversion lines for all the elements studied was made from a reasonably accurate value of the field as measured with a previously standardized coil and fluxmeter. The accurate x-ray energy level values were then used to calculate the value of the field which brought the transition energies derived from the *K*, *L*, *M*, and *N* conversions into agreement. Second, the accurately known 80-kev gamma-ray of I<sup>131</sup> was used as a standard for calibrating the magnetic field. Du Mond<sup>4</sup> and his co-workers have measured the gamma-rays from I<sup>131</sup> using a double crystal spectrometer. They report a value of  $80.133 \pm 0.005$  kev for the low energy gamma-ray. A sample of radioactive separated isotope I<sup>131</sup> obtained from Oak Ridge was used as a source in the spectrograph. Three conversion electron lines found were assigned to *K*, *L*, and *M* conversions of the 80-kev gamma-ray. Using Du Mond's value for the gamma-ray energy, the measured radii of curvature of the three electron lines, and the x-ray data for the critical absorption energies, a value was assigned to the magnetic field. The values of the magnetic field obtained by the two methods agreed to within 0.1 percent and the weighted mean value of  $98.3 \pm 0.1$  gauss was adopted for the calculation of the electron energies.

The field of the spectrograph magnet was uniform to within 0.2 percent except at points closer than 5 cm from the edge of the pole pieces, which measured  $30.5 \times 50.8$  cm. The maximum useful radius of the spectrograph was 20 cm. The maximum value of the product of field strength and electron radius of curvature obtainable was then 1966 gauss-cm, corresponding to an electron energy of 273 kev. Although electrons of energy as low as 5 kev were focused on the detector, the lower energy limit of the spectrograph was probably about 20 kev because of the low sensitivity of the film to electrons of energy less than 20 kev.

## 2. TRANSITION ENERGIES

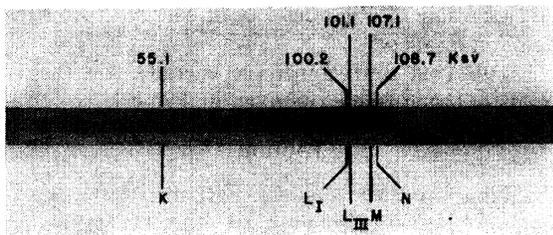
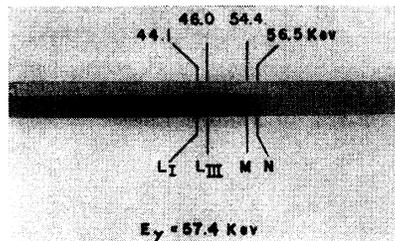
### Cesium

Neutron irradiation of cesium, which has but one stable isotope of mass number 133, produces a 3-hour period which is isomeric with a long period of about 2 years. Goldhaber and Muehlhause<sup>5</sup> reported for the 3-hour period strong *K<sub>α</sub>* x-rays of cesium as well as conversion electrons which they assumed to be *K* electrons of a transition of about 160 kev. Peacock, Jones, and Overman<sup>6</sup> reported a highly converted gam-

<sup>4</sup> Lind, Brown, Klein, Muller, and Du Mond, Phys. Rev. **75**, 1544 (1949).

<sup>5</sup> M. Goldhaber and C. O. Muehlhause, Phys. Rev. **74**, 1904 (1948).

<sup>6</sup> Peacock, Jones, and Overman, Plutonium Project Report, Mon. N-432, 56 (1947).

FIG. 2. Internal conversion electrons of Dy<sup>166</sup> (1.3 min.).FIG. 3. Internal conversion electrons of Ir<sup>192</sup> (1.5 min.).

ma-ray of  $150 \text{ keV} \pm 30 \text{ keV}$  going to the lower state of Cs<sup>134</sup> (2 yr.).

A spectrogram of the 3-hour period of cesium showed three electron lines due to a single gamma-ray of energy  $128 \pm 0.5 \text{ keV}$  which converts in the *K*, *L*, and *M* shells. Table I gives the energies of the internal conversion electrons. The previously reported values would be in good agreement with our value if *L* rather than *K* electrons had been assumed. An examination of the critical absorption energies<sup>7</sup> for both cesium and barium shows that the *K*–*L* electron energy difference and the *L*–*M* electron energy difference fit the accurate x-ray absorption difference for these shells for cesium but do not fit the same shell differences for barium. This confirms the critical absorption experiments of Goldhaber and Muehlhause,<sup>5</sup> showing that the transition is an isomeric one in cesium and not a transition following a beta-ray which would require that the transition be in the element barium.

### Cobalt

The 10.7-minute period in cobalt produced by neutron irradiation of cobalt metal has been investigated by Deutsch, Elliott, and Roberts.<sup>8</sup> They report that at least ninety percent of the disintegrations are by an isomeric transition of energy 56 keV, presumably to the five-year level. The remaining ten percent of the disintegrations are by a beta-ray of 1.25 MeV, followed by a single gamma-ray of energy 1.50 MeV. They studied the beta-ray spectrum with a spectrometer having a thin window counter and found a peak of conversion electrons at  $48.5 \pm 2 \text{ keV}$ . The *K* and *L* peaks were not resolved because a thick source was used. Assigning the conversion electrons to conversion mostly in the *K*-shell, they obtained a value of  $56 \pm 3 \text{ keV}$  for the energy of the gamma-ray.

In the present work, a spectrogram of the 10.7-minute period of cobalt showed three conversion electron lines which were assigned to *K*, *L*, and *M* conversions of a single gamma-ray of energy  $58.9 \pm 0.5 \text{ keV}$ . The electron energies are shown in Table I.

### Columbium

The 6.6-minute period in columbium can be produced by slow neutron irradiation of columbium.<sup>9</sup> Goldhaber, Muehlhause, and Turkel<sup>10</sup> found that very pure Cb exposed to slow neutron irradiation emitted strong *K* x-rays. By critical absorption they showed these x-rays to be characteristic *K* radiations of Cb which implies that they arise from an isomeric transition in columbium. The internal conversion electrons were compared with the known internal conversion electrons from Co<sup>60</sup> (10.7-min. period). A value of 36.5 keV was obtained; adding the Cb *K* work function of 19.0 keV gave 55.5 keV for the energy of the isomeric transition. Beta-rays of energy 1.3 MeV were also found. No unconverted gamma-rays were found, indicating that practically all of the gamma-rays are internally converted.

A spectrogram of the 6.6-minute internal conversion electrons was obtained using a thin foil ( $\sim 2 \text{ mg/cm}^2$ ) of columbium metal irradiated with slow neutrons. It was necessary to make 100 bombardments for a sufficient exposure. Three internal conversion electron lines were observed of energies 22.5, 39.0, and 40.8 keV.

TABLE II. Relative intensities of electron lines.

Element and half-life	Electron shell	Electron energy (keV)	Relative intensity (arbitrary units)
Cesium (3 hr.)	<i>K</i>	92.0	21.7
	<i>L</i>	123.0	33.8
	<i>M</i>	127.0	6.3
Cobalt (10.7 min.)	<i>K</i>	51.1	29.5
	<i>L</i>	58.2	6.5
	<i>M</i>	58.7	2.1
Columbium (6.6 min.)	<i>K</i>	22.5	1.4
	<i>L</i>	39.0	4.5
	<i>M</i>	40.8	1.6
Dysprosium (1.3 min.)	<i>K</i>	55.1	1.8
	<i>L<sub>I</sub></i>	100.2	14.3
	<i>L<sub>III</sub></i>	101.1	9.4
	<i>M</i>	107.1	6.3
	<i>N</i>	108.7	3.0
Iridium (1.5 min.)	<i>L<sub>I</sub></i>	44.1	26.0
	<i>L<sub>III</sub></i>	46.0	16.2
	<i>M</i>	54.4	10.1
	<i>N</i>	56.5	3.2

<sup>7</sup> A. H. Compton and S. K. Allison, *X-Rays in Theory and Experiment* (D. Van Nostrand Company, Inc., New York, 1935).

<sup>8</sup> Deutsch, Elliott, and Roberts, *Phys. Rev.* **68**, 193 (1945).

<sup>9</sup> M. Goldhaber and W. J. Sturm, *Phys. Rev.* **70**, 111A (1946).

<sup>10</sup> Goldhaber, Muehlhause, and Turkel, *Plutonium Project Report*, CF-3574 (July, 1946).

TABLE III. Multipole order of the radiation.

Element	Half-life	Transition energy (kev)	Multipole order	
			From lifetime —energy relation	From K/L ratio
Cs	3 hr.	128.0	2 <sup>4</sup> or 2 <sup>5</sup>	2 <sup>4</sup> or 2 <sup>5</sup>
Co	10.7 min.	58.9	2 <sup>4</sup>	2 <sup>4</sup>
Cb	6.6 min.	41.5	2 <sup>4</sup>	2 <sup>4</sup>
Dy	1.3 min.	109.0	2 <sup>4</sup>	2 <sup>4</sup>
Ir	1.5 min.	57.4	2 <sup>4</sup>	—

These were assigned to *K*, *L*, and *M* conversions of a single gamma-ray of energy  $41.5 \pm 0.5$  kev. The *L* conversion line is undoubtedly what Goldhaber *et al.*, interpreted as the *K* line in their aluminum absorption experiment. The electron energies for columbium are given in Table I.

### Dysprosium

When the element dysprosium is subjected to slow neutron bombardment, two periods are observed. The 2.6-hour activity was first reported by Hevesy and Levi<sup>11</sup> and was confirmed by Pool and Quill<sup>12</sup> and by Inghram, Shaw, Hess, and Hayden.<sup>13</sup> The 1.25-minute *K*-converted gamma-activity was initially reported by Flammersfeld.<sup>14</sup> Inghram *et al.*, showed that both of these activities are produced by neutron irradiation of the stable isotope Dy<sup>164</sup> and that the 1.25-minute activity is isomeric with the 2.6-hour activity.

For the 1.25-minute activity Flammersfeld has reported conversion electrons of energy 130 kev as measured by absorption in aluminum. He interpreted these as *K* electrons from which he obtained an energy of about 180 kev for the isomeric transition. Hole,<sup>15</sup> using a beta-ray spectrometer, observed an electron line at 93 kev which he interpreted as an *L* conversion electron line.

A spectrogram of the 1.25-minute activity, reproduced in Fig. 2, was obtained by making fifteen bombardments to give an accumulated exposure on one film. There were five conversion electron lines, which were assigned to a single gamma-ray of energy  $109.0 \pm 0.5$  kev which converts in the *K*, *L*<sub>I</sub>, *L*<sub>III</sub>, *M*, and *N* shells of dysprosium.

Seaborg and Perlman<sup>16</sup> report for the 2.6-hour activity of dysprosium four gamma-rays of energy 91, 370, 780, and 1100 kev. A spectrogram of the 2.6-hour activity showed six conversion electron lines, five of which were assigned to *K*, *L*<sub>I</sub>, *L*<sub>III</sub>, *M*, and *N* conversions of a single gamma-ray of energy  $87.8 \pm 0.7$  kev. The sixth line was not assigned, but is probably a *K*

<sup>11</sup> G. Hevesy and H. Levi, *Nature* **136**, 103 (1935).

<sup>12</sup> M. L. Pool and L. L. Quill, *Phys. Rev.* **53**, 437 (1938).

<sup>13</sup> Inghram, Shaw, Hess, and Hayden, *Phys. Rev.* **72**, 515 (1947).

<sup>14</sup> A. Flammersfeld, *Zeits. f. Naturforschung* **1**, 190 (1946).

<sup>15</sup> N. Hole, *Arkiv. f. Mat. Astr. o. Fys.* **36A**, paper 2 (1948).

<sup>16</sup> G. T. Seaborg and I. Perlman, *Rev. Mod. Phys.* **20**, 585 (1948).

conversion line of a gamma-ray of energy greater than three hundred kev.

The electron and transition energies for the two periods of dysprosium are given in Table I.

### Iridium

Iridium has three neutron activated periods. The 1.5-minute period is isomeric with the seventy-two day period and both are assigned to Ir<sup>194</sup>. Goldhaber Muehlhause, and Turkel<sup>17</sup> report for the 1.5-minute period two gamma-rays of energy approximately 60 kev and 30 kev as well as strong *L* x-rays of iridium. They were unable, however, by critical absorption to show that there is a unique gamma-ray in the region of about thirty kev.

A spectrogram of the 1.5-minute activity using some of the same sample of iridium as used by Goldhaber *et al.*, was made and is shown in Fig. 3. Four conversion electron lines were seen which were assigned to a single gamma-ray of energy  $57.4 \pm 0.5$  kev which converts in the *L*<sub>I</sub>, *L*<sub>III</sub>, *M*, and *N* shells of iridium. A second spectrogram was made using twice as long an exposure on the film to try to find lower energy electrons supposedly from conversion of the  $\sim 30$ -kev gamma-ray. The results obtained were the same as for the first exposure, indicating no discrete lower energy gamma-rays. The electron energies for iridium are shown in Table I. The following interpretation has been suggested.<sup>18</sup> The transition from the 1.5-minute metastable state to the ground state can take place either by a 2<sup>4</sup>-pole isomeric transition or by a competitive "double photon" emission.

A spectrogram of the nineteen-hour activity showed no conversion electron lines in the energy range of the spectrograph.

### Thulium

The 120-day period in thulium was produced by neutron irradiation of thulium oxide powder. This period is reported to have a beta-ray spectrum of 1-Mev maximum energy and no gamma-rays.<sup>16</sup> Saxon<sup>3</sup> recently studied this period using a 180° focusing magnetic spectrometer and found superimposed on the beta-ray spectrum three internal conversion electrons of a single gamma-ray of energy<sup>19</sup> 85.5 kev.

Using a sample of irradiated thulium obtained from Mr. Saxon a spectrogram was made in the permanent magnet spectrograph. Five electron lines were clearly seen, which were assigned to *K*, *L*<sub>I</sub>, *L*<sub>III</sub>, *M*, and *N* conversion electrons of a single gamma-ray of energy  $84.8 \pm 0.5$  kev. The electron energies are given in Table I. The *L*<sub>I</sub> and *L*<sub>III</sub> conversion electron lines were clearly resolved in the spectrogram as two separate

<sup>17</sup> Goldhaber, Muehlhause, and Turkel, *Phys. Rev.* **71**, 372 (1947).

<sup>18</sup> M. Goldhaber (private communication).

<sup>19</sup> See also R. L. Graham and D. H. Tomlin, *Nature* **164**, 278 (1949) who find for the gamma-ray an energy of 82.7 kev.

lines although the energy difference between them is only 1.1 kev.

### 3. RELATIVE INTENSITIES OF ELECTRON LINES AND MULTIPOLE ORDER OF THE RADIATION

Relative intensities of the conversion electron lines were measured for the five isomeric transitions studied. Corrections to the observed densities were made for fog, density-exposure, and for differences in film sensitivity for electrons of different energies. The resulting corrected values of relative intensities of electron lines is given in Table II and are to be considered only as approximate values.

For each of the five isomeric transitions the multipole order of the radiation was obtained by two independent methods. First, the half-life period of each element was checked with the reported value by observing the decay of the radiations from samples bombarded in the pile using an ion-chamber detector. These values of the half-life periods and the spectrograph values of the energies of the transitions were used together with Wiedenbeck's<sup>20</sup> curves to determine the multipole order of the radiation, assuming electric multipole radiation.

<sup>20</sup> M. L. Wiedenbeck, *Phys. Rev.* **69**, 567 (1946).

Second, the ratio of the intensities of the *K* and *L* conversion electrons were used together with the theoretical curves of Hebb and Nelson<sup>21</sup> to find the multipole order of the radiation for each of the four isomeric transitions where both *K* and *L* conversions were observed.

Table III gives the values obtained for the multipole order assignments as well as a summary of the transition energy data. Axel and Dancoff<sup>22</sup> used some of these data in their recent analysis of isomeric transitions and reached similar conclusions.

The author wishes to express his deep appreciation of the advice and assistance generously given to him by the following: Professor Newell S. Gingrich, University of Missouri, and Professor M. Goldhaber, University of Illinois consultant to the Argonne National Laboratory, for suggesting the problem and for guidance throughout the investigation; Messrs. E. der Mateosian, R. D. Hill, C. O. Muehlhause, and S. B. Burson for many helpful discussions. To the O. M. Stewart Fund of the University of Missouri appreciation is expressed for financial support granted.

<sup>21</sup> M. H. Hebb and E. Nelson, *Phys. Rev.* **58**, 486 (1940).

<sup>22</sup> P. Axel and S. M. Dancoff, *Phys. Rev.* **76**, 892 (1949).

## Cosmic-Ray Intensity Following a Solar Flare\*

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(Received January 30, 1950)

Continuous records of the cosmic-ray intensity measured by a heavily shielded, high pressure ion chamber, were obtained during and following an intense solar flare (of importance 3+) and a subsequent intense magnetic storm. No increase in *C-R* intensity was observed at or near the time of the flare, such as have been observed by others on similar occasions. At the peak of the magnetic storm there was a decrease of about 1.5 percent in *C-R* intensity, with full recovery requiring 2 or 3 days. Unshielded *C-R* telescope readings of low accuracy are included. These indicate that there were no prolonged alterations in the intensity (without shielding) of magnitude much greater than 14 percent associated with either the flare or the storm.

### 1. INTRODUCTION

IN recent years there have been reported instances of changes in cosmic-ray intensity apparently associated with particular solar flares. For example, Forbush<sup>1</sup> reported that upon examination of 10 years of continuous records with ion chambers shielded by 11 cm Pb, three unusual increases were noted. These occurred on February 28 and March 7, 1942, and July 25, 1946. The first two consisted of sharp increases of about 7 percent which began within 18 minutes after radio fade-outs which were presumed to indicate solar flares.

\* The experimental work was supported in part by the ONR.  
<sup>1</sup> S. E. Forbush, *Phys. Rev.* **70**, 771 (1946); 1942 increases reported also by I. Lange and S. E. Forbush, *Terr. Mag.* **47**, 331 (1942).

The third represented a sharp increase of about 15 percent beginning one hour after the commencement of a solar flare and fade-out. The peaks were very sharp, being represented on the graphs by only one or two points representing bi-hourly means, with the entire increase above indicated statistical deviations being represented by about 3 to 6 points. After a very few hours, the intensity returned to normal and not long afterward it dropped to some 7 or 8 percent subnormal in the first and third instances. The large subnormal dips followed a few hours after the sudden commencement of magnetic storms, and the upward return toward normal was slow, extending over several days. In the third case, the magnetic storm began 26.8 hours after the solar flare. The reliability of the observations of the

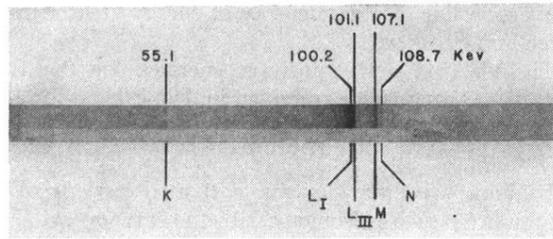


FIG. 2. Internal conversion electrons of  $Dy^{166}$  (1.3 min.).

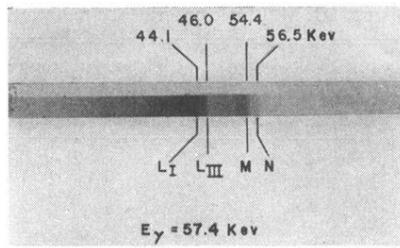


FIG. 3. Internal conversion electrons of Ir-192 (1.5 min.).