The Measurement and Interpretation of the K Auger Intensities of Sn^{113}, Cs^{137}, and Au^{198}

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Auger electron intensity measurements have been made on Cs^{137}, Au^{198}, and Sn^{113} with a magnetic lens spectrometer which is briefly described. K Auger yields for Ba and Hg of 0.130 ± 0.007 and 0.056 ± 0.008, respectively, were obtained. The K Auger electrons were resolved into three groups, K–LL, K–LX, and K–XY, according as two, one, or no L electrons were involved. The ratios of the intensities of the latter two to the K–LL were found to be for In 0.417 ± 0.016 and 0.076 ± 0.008; for Ba 0.40 ± 0.04 and 0.08 ± 0.02. The measured ratio of K–LX to K–LL for Hg was 0.71 ± 0.15. The relationship of these results to theory is discussed. A summary of measurements of K fluorescence yield and K Auger line intensity ratios is given. The Auger spectrum from Au^{198} indicates that K capture to Pt^{198} cannot occur in more than 0.5 percent of the disintegrations. A crude estimate of the L to K capture ratio for Sn^{113} indicates that it is not abnormally high.

I. INTRODUCTION

IN the decay of radioactive nuclei, vacancies in the K electron shell are frequently created by internal conversion and by K capture. It is often desirable to determine the combined probability of these processes by counting the number of K vacancies produced per disintegration. As is well known,[1] a K vacancy is always filled by an electron from a shell of higher energy. The resulting energy can either be emitted as electromagnetic radiation (K x-rays), or be transferred to an electron in a shell of higher energy and eject it from the atom (K Auger electron).

If the K fluorescence yield w_K is known, a count of the empty shells can be made by a count of either the K x-rays or the K Auger electrons. The K fluorescence yield is defined as the number of K x-rays emitted per K vacancy. In the past, most of the measurements of vacancies in the K shells have been made by means of the K x-rays in spite of the difficulty in making absolute x-ray intensity measurements, because the K Auger electrons cannot penetrate the windows of most counters and ion chambers. A further difficulty is the lack of accurate knowledge of fluorescence yield, particularly for atomic number greater than fifty. This study attempts to increase the accuracy of our knowledge of the fluorescence yield for elements with atomic number greater than fifty, and to point out the possible usefulness of K Auger electron spectra obtained in a magnetic spectrometer with a very thin window.

II. EXISTING KNOWLEDGE OF THE FLUORESCENCE YIELD

A. Theoretical

Any complete and accurate theory of the Auger effect would be based on a proper theory of quantum electrodynamics, since the effect involves the interaction of several electrons.[2] Such a theory in a satisfactory state does not exist at present. Approximate theoretical treatments of the Auger effect, however, have been made by several workers. The first considerations were by Wentzel,[3] who found that the K Auger transition probability γ_A should be independent of the atomic number Z. Since the x-ray transition probability γ_X is approximately proportional[1] to Z^4, the K Auger yield should be approximately

$$a_K=\gamma_A/(\gamma_A+\gamma_X)=C_1/(C_1+C_2Z^4)=1/(1+bZ^4), \quad (1)$$

and the fluorescence yield

$$w_K=1-a_K=\gamma_X/(\gamma_A+\gamma_X)$$
$$=C_2Z^4/(C_1+C_2Z^4)=Z^4/(a+Z^4). \quad (2)$$

Wentzel did not make any numerical calculations, but Pincherle,[4] Burhop,[5] and Ramberg and Richtmyer[6] made use of the expressions derived by him to calculate values for some of the Auger transitions involving an

TABLE I. Transition probabilities per unit atomic time ($\times10^3$).[a]

Transition	Massey and Burhop Gold[r,s]		Burhop Silver[n,s]		Ramberg and Richtmyer Gold[n,f]		Pincherle[n,u]	
K–L_IL_I	1.74		2.54		2.02		2.50	
K–L_IL_{II}	9.52		2.88		2.36			
		18.73		8.65		6.82		8.81
K–L_IL_{III}	9.21		5.77		4.46			
K–$L_{II}L_{II}$			2.43					
				8.91		1.41		1.88
K–$L_{II}L_{III}$			6.48					
K–$L_{III}L_{III}$			8.20		17.88		22.44	
K–LX							25.7	
K–XY							2.70	

[a] Unit atomic time is the period of the electron of the first Bohr orbit of hydrogen. r—relativistic; n—nonrelativistic; s—screened hydrogen-like wave functions; u-unscreened hydrogen-like wave functions; f—Fermi-Thomas statistical model for potential field.

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[1] A. H. Compton and S. K. Allison, *X-Rays in Theory and Experiment* (D. Van Nostrand Company, Inc., New York, 1935), second edition, p. 482.
[2] N. F. Mott and I. N. Sneddon, *Wave Mechanics and Its Application* (Oxford University Press, London, 1948), pp. 338 and 342.
[3] G. Wentzel, Z. Physik **43**, 524 (1927).
[4] L. Pincherle, Nuovo cimento **12**, 81 (1935).
[5] E. H. S. Burhop, Proc. Roy. Soc. (London) **A148**, 272 (1935).
[6] E. G. Ramberg and F. K. Richtmyer, Phys. Rev. **51**, 913 (1937).

initial K shell vacancy. Table I lists these values as well as those of Massey and Burhop[7] discussed below. The first three calculations used nonrelativistic wave functions for the electrons. Pincherle used unscreened hydrogen-like wave functions in a Coulombian field; in this approximation the transition probability is completely independent of Z. He has made the most complete set of calculations of probabilities for the K Auger electrons, considering interactions of electrons in the K, L, M, and N shells. In the spectrometer used by the authors, the K Auger electrons are resolved into three groups or lines, which may be designated as the K–LL, K–LX, and K–XY Auger lines. Here X and Y are used to denote M, N, etc., shell electrons. Thus, the K–LX line is due to K–LM, K–LN, etc., Auger electrons, and the K–XY line is due to K–MM, K–MN, K–NN, etc., Auger electrons. It will be interesting to compare the observed ratios of intensities of these three lines to those obtained from the calculated probabilities of Pincherle. These calculated ratios are K–LL:K–LX:K–XY::1.00:0.716:0.103.

Burhop, using the more accurate screened hydrogen-like wave functions with effective nuclear charges for silver as determined by Slater,[8] has calculated the transition probabilities only for the K–LL interactions. Ramberg and Richtmyer used the still more accurate Fermi-Thomas statistical model for the potential field in calculating the K–LL probabilities for gold. The above three works are in reasonable agreement for the total probability of the K–LL interactions, the only ones common to the three calculations. The relative values of the probabilities of the various K–LL transitions are closely the same for Pincherle, and Ramberg and Richtmyer, though the absolute values of the latter are about twenty-five percent smaller. In a comparison of Burhop's and Pincherle's values, it is notable that the K–$L_{III}L_{III}$ probability of Burhop is only slightly more than one-third that of Pincherle, while his K–$L_{II}L_{II}$ and K–$L_{II}L_{III}$ values are several times greater than Pincherle's.

Relativistic calculations have been made by Massey and Burhop[7] for the K–L_IL_I, K–L_IL_{II}, and K–L_IL_{III} transition probabilities in gold and the K–L_IL_{II} probabilities in silver. The theoretical method, due to Møller,[9] on which Massey and Burhop base their work, considers only the two electrons directly involved in the transition. This is true of the nonrelativistic calculations also. In this method,[2] the charge and current densities associated with the electron transition to the K vacancy are written with the aid of the Dirac theory. The retarded potentials of the electromagnetic field due to the transition are then calculated from these densities. These potentials are then used as the perturbation in a first-order perturbation calculation of the transition probability for the ejection of another orbital electron. Using screened hydrogen-like wave functions, they find that the use of relativity increases the calculated Auger probability. For the transitions considered the increase is twenty percent for silver and almost one hundred percent for gold. In addition they find that the use of relativity decreases the calculated x-ray transition probability by a factor of 0.75 in gold so that the Auger yield for gold is slightly more than double the nonrelativistic value of 0.03. Using the K–LL Auger yields of Burhop, the relative values of the K–LL, K–LX, and K–XY yields of Pincherle, and corrections due to relativity based on their own calculations, Massey and Burhop constructed a theoretical curve of K Auger yield as a function of Z. Thus, the best theoretical curve available consists of a combination of several incomplete and approximate calculations. Values taken from this curve are $a_K=0.06$ for mercury, $a_K=0.15$ for barium, and $a_K=0.216$ for indium.

B. Experimental

Many workers have measured K fluorescence yields for low atomic number elements (Z less than fifty) by observation of incident and fluorescent x-ray intensities. The summaries of Arends[10] in 1935 and Backhurst[11] in 1936 cover the contemporary data very well. Backhurst points out that some persons have confused the K fluorescence yield with the number of K x-rays per orbital (K, L, M, etc.) vacancy. This confusion has occurred in summaries that include the results of Harms,[12] Balderston,[13] and Martin,[14] who report the latter quantity in their papers. The summaries of Compton and Allison,[1] Massey and Burhop,[7] Steffen *et al.*,[15] and Tellez-Plasencia[16] report the values of Harms, Balderston, and Martin as w_K. A summary by Stephenson[17] reports the work of Martin correctly, but omits the results of Harms, Locher,[18] and others, while Locher gives more weight to Harms' and Compton's[19] results than to Martin's. There is some reason for omitting the results of Berkey.[20] He finds a maximum value for w_K at $Z=42$. Such a maximum is not supported by any other experimental data or by theory. Backhurst suggests that this maximum is due to the fact that the incident x-rays used by Berkey may not have been monoenergetic. None of these summaries includes the several measurements of K yields made since 1940. These measurements include the only ones made for elements of high atomic number.[20a]

[7] H. W. S. Massey and E. H. S. Burhop, Proc. Roy. Soc. (London) **A153**, 661 (1936).
[8] J. C. Slater, Phys. Rev. **36**, 57 (1930).
[9] C. Møller, Z. Physik **70**, 780 (1931).
[10] E. Arends, Ann. Physik **22**, 281 (1935).
[11] I. Backhurst, Phil. Mag. **22**, 737 (1936).
[12] M. I. Harms, Ann. Physik **82**, 87 (1927).
[13] M. Balderston, Phys. Rev. **27**, 696 (1926).
[14] L. H. Martin, Proc. Roy. Soc. (London) **A115**, 420 (1927).
[15] R. Steffen *et al.*, Helv. Phys. Acta **22**, 167 (1949).
[16] H. Tellez-Plasencia, J. phys. et radium **10**, 14 (1949).
[17] R. Stephenson, Phys. Rev. **51**, 637 (1937).
[18] G. L. Locher, Phys. Rev. **40**, 484 (1932).
[19] A. H. Compton, Phil. Mag. **8**, 961 (1929).
[20] D. K. Berkey, Phys. Rev. **45**, 437 (1934).
[20a] *Note added during publication:* A recent book by Burhop, *The Auger Effect and Other Radiationless Transitions* (Cambridge

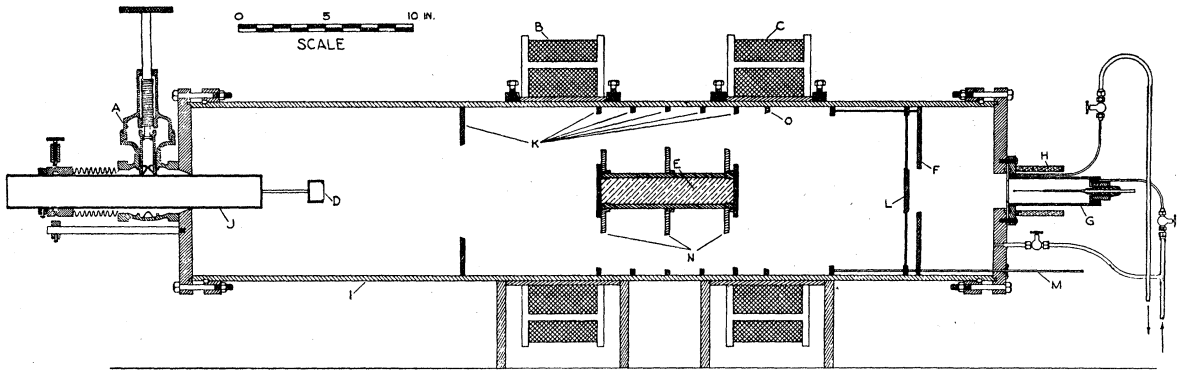

FIG. 1. Cross section of the lens spectrometer. (A) Vacuum lock, (B, C) lens coils, (D) source, (E) lead absorber, (F, L) ring baffles, (G) counter, (H) counter shield, (I) aluminum vacuum tank, (J) source holding tube, (K) anti-scattering baffles, (M) ring baffle position adjusting rod.

The experimental values support in a qualitative way the theoretical variation of a_K (or w_K) with atomic number, but none of the curves yet suggested, theoretical or semi-empirical, is in satisfactory agreement with all the available data. The relation

$$w_K = 0.957Z^4(0.984\times 10^6 + Z^4)^{-1}, \qquad (3)$$

given by Arends[10] and recommended by Tellez-Plascencia,[16] seems to give the best fit of those that have been proposed. A new and, it is hoped, complete and accurate summary is given in Sec. V.

In addition to the values of w_K, several measurements of the relative intensities of the various K Auger lines have been reported in the lireature. Ference,[21] using a magnetic spectrometer and Geiger-Mueller counter, studied the K Auger electrons from germanium. He reported the ratio K–LL:K–LX::1.00:0.31. He did not resolve the K–LL Auger electrons well enough to allow an accurate estimate of the ratio of the K–LL sublevels, but the shape of the line indicated three groups of electrons with the highest energy group being the most intense. He gave the ratios of intensities of these groups, to be taken as an order of magnitude only, as 1.00:0.26:0.26. In 1950 Bergstrom and Thulin,[22] using the same technique as the present authors, obtained the ratio K–LL:K–LX::1.00:0.48, for Cs^{133}. Steffen *et al.*,[15] using two isotopes of platinum, obtained average ratios of K–LL:K–LX:K–XY::1.00:0.86:0.025. Huber *et al.*[23] obtained the ratios K–LL:K–LX:K–XY::1.00:0.49:0.054 for silver and 1.00:0.44:0.056 for cadmium.[23a]

III. EXPERIMENTAL PROCEDURE

A. The Magnetic Lens Spectrometer

The lens spectrometer used in this work was designed for medium resolution and accurate relative intensities over a wide range of energies. A cross section of the vacuum chamber and coils is shown in Fig. 1, from which the many conventional details can be inferred. The chamber is unusual in that it is a machined aluminum cylinder (o.d. $10\frac{1}{4}$ in, i.d. $9\frac{1}{2}$ in.) with no welds whatever. The vacuum line is connected by an aluminum tube with a tapered pipe thread. Immediately before being screwed to a binding tightness both threads were painted with glyptal[24] lacquer. The end plates are each bolted to an aluminum collar, which fits snugly on the tube and is held fixed against a split ring which fits into a groove machined into the aluminum tube and projects $\frac{1}{8}$ in. above the surface of the tube.

The accuracy of intensity measurements is insured by the absence of any iron without four feet of the spectrometer, by coils which compensate for the earth's magnetic field to better than 0.1 percent,[25] by maintenance of a vacuum of 10^{-5} mm of mercury or better, and by the use of counter window thicknesses down to 10 μg cm^{-2}. Both Nylon and collodion windows supported by Lectromesh[26] have been used.

A gas flow ($\frac{2}{3}$ argon and $\frac{1}{3}$ ethylene) is continuously maintained through the counter at 4- to 6-cm pressure. It has been verified that at 6-cm pressure the counting rate is independent of electron energy up to 1.7 Mev. The counter voltage is adjusted to keep constant the pulse height as observed in an oscilloscope. It was found by observation of a constant strength source that this procedure maintained constant counter sensitivity even though, because of aging or change in gas pressure, the operating voltage varied as much as 10 percent.

University Press, Cambridge, England, 1952), pp. 44–51, has a more complete and more accurate summary. However, the values of Harms quoted are not for w_K as was pointed out by Backhurst, nor are those of Locher his preferred values. The summary in Section V attempts to rectify these and other minor errors and has a few additional references, some old and some published since 1950.

[21] M. Ference, Phys. Rev. **51**, 727 (1937).

[22] I. Bergstrom and S. Thulin, Phys. Rev. **79**, 539 (1950).

[23] O. Huber *et al.*, Helv. Phys. Acta **25**, 3 (1952).

[23a] *Note added during publication:* We had previously overlooked the work of Ellis and Flammersfeld (see references in Section V), who have measured the relative intensities of several of the lines for Bi (Z=83) using 180° magnetic spectrometers. From Ellis this K–LX to K–LL ratio is greater than 0.4 and from Flammersfeld it is at least 0.57.

[24] 1201 Glyptal lacquer, made by the General Electric Company.

[25] S. K. Haynes and J. W. Wedding, Rev. Sci. Instr. **22**, 97 (1951).

[26] Lektromesh, C. O. Jelliff Manufacturing Company, Southport, Connecticut.

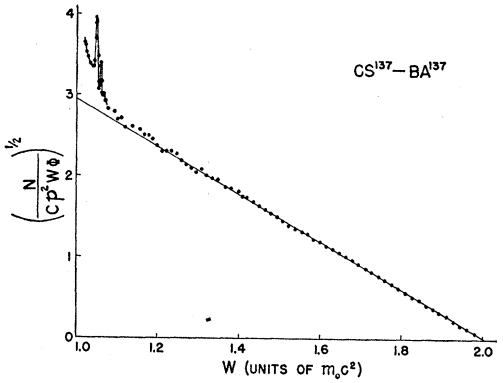

FIG. 2. Fermi-Kurie plot of the spectrum of the Cs^{137} source after subtraction of the 1.2-Mev beta-ray.

Each of the lens coils contains 1250 turns of 0.050 $\times$0.150 in. double Formex wire wound between sides composed of $\frac{3}{8}$ in. square copper tubing. A layer of tubing is also imbedded in the middle of each coil. Each coil is mounted on the main spectrometer tube by six adjustable screws. After the best possible visual alignment has been achieved, final adjustment is made with a low energy electron beam traversing the spectrometer without baffles. The compensating coil current is first adjusted so that the electron beam position is independent of beam energy.[25] Each lens coil is then adjusted individually until, as its current is varied, foci of all orders fall at the center of a fluorescent screen which replaces the counter.

The source of current for the lens coils is a 250-v, 6-kw dc generator controlled by an amplidyne. The input to the amplidyne is the amplified error signal between the desired current as set up on a type K potentiometer and the actual current as represented by the potential difference across a standard resistor. Negative feedback in the amplifier and a large LC filter keep current fluctuations of all frequencies to 0.1 percent or less.

The method of calculation of the electron trajectories from a point source in this spectrometer and the method of calculation of the ring baffle positions have already been described.[27] The theoretical resolution for a point source and a transmission (acceptance angle/4π) of 0.015 is $\Delta p/p=0.017$. In practice a resolution of 0.024 was obtained for the 3-mm diameter source of Au^{198} reported below. The run on In^{113} was made with different source diameter, coil geometry, and baffle arrangement, and the run on Cs^{137} with a different baffle arrangement which resulted in poorer resolution for these runs. Under optimum conditions the resolution for a source 6 to 8 mm in diameter is 0.030 for a transmission of 0.015.

[27] J. F. Perkins and A. W. Solbrig, Jr., Rev. Sci. Instr. **22**, 173 (1951).

B. Source Preparation

In order to measure intensities of electrons with energies between 20 and 80 kev one must have very thin uniform sources mounted on very thin backings. Vacuum distillation and electroplating are probably the only methods capable of giving sufficient uniformity. The former method was used for Cs^{137} and Au^{198} and could have been used for Sn^{113} if sufficient activity had been available. The Sn^{113} source was electroplated in order to conserve material.

The vacuum distillation chamber consists of a brass base covered with a "bell jar" about five inches high made from two-inch glass tubing. A $\frac{1}{16}$-inch rubber gasket with vacuum grease provides the seal between the base and the glass. Atmospheric pressure is sufficient to provide a good seal. The radioactive material is placed on a tantalum strip filament 0.001$\times$0.050 in., which is supported by the filament leads through the base. The source film holder is mounted and centered 0.020 in. behind a radiation shield which is $\frac{1}{4}$ in. above the center of the filament. The source backing for the cesium and gold was of Formvar of thickness approximately 75 μg cm^{-2} made conducting by a thin coat of evaporated aluminum.

For the cesium source a few drops of cesium chloride solution were dried on the filament. After evacuation the filament was heated to approximately 1000°C for one minute; the resulting source had an average thickness of 7 μg cm^{-2} and a diameter of 3 mm. In the case of the gold source the strip of gold foil received from Oak Ridge was simply laid on the filament and after evacuation heated to about 1400°C for one minute. The resulting source had a thickness of about 20 μg cm^{-2} and a diameter of 3 mm. The uniform density of autoradiographs and the lack of low energy tails on the lines in the spectrometer indicated, respectively, that the sources were uniform and thin.

The tin, enriched in Sn^{112} prior to irradiation,[28] was received in the form of 1 mg of SnO_2. It was reduced to metallic tin by being heated to approximately 1000°C in hydrogen gas. The metallic tin was dissolved in a hydrochloric acid plating solution by a procedure outlined by Sand.[29] A Formvar film of thickness less than 10 μg cm^{-2} coated with about 75 μg cm^{-2} of evaporated copper was mounted on an aluminum ring 1 in. in diameter. A piece of platinum gauze was placed so that a flat circular section about 0.5 cm in diameter was held $\frac{1}{8}$ in. above the film. After a positive potential of 1.12 volts with respect to the film had been applied, one to three drops of plating solution were dropped through the gauze so that they formed by surface tension a roughly cylindrical plating cell. The voltage was immediately raised to 1.9 volts. Plating continued for

[28] We are indebted to Dr. G. E. Boyd of Oak Ridge National Laboratory for the sample of Sn^{113} used.

[29] H. J. S. Sand, *Gravimetric Electrolytic Analysis* (Blackie and Son, Ltd, London and Glasgow, 1940), p. 72 (description of the method of A. C. Penney).

about 8 min at 1.5 ma. A uniform source of Sn^{113} having a thickness of much less than 0.1 mg cm^{-2} was plated out. The solution was neutralized with hydrazine hydroxide before the potential was disconnected. The source was washed by twenty additions and removals of two or three drops of distilled water. It should be emphasized that the potential must never be completely removed from the cell as long as the solution is acid. The acid solution demolishes the copper film rapidly and also tends to redissolve the tin already plated.

IV. EXPERIMENTAL RESULTS

A. Cs^{137}—Ba^{137}

The observations made were the time intervals required to accumulate a pre-set number of counts for different settings of the lens-coil current. The current was varied in steps of 0.2 percent in the region of the Auger and conversion electron lines, and in steps of 0.5 to 1.0 percent over the continuous spectrum. Eight runs were made across the desired current range alternately in the direction of increasing and decreasing current. A total of 3200 or more counts were obtained at each current setting. The current stability and counter sensitivity were checked at intervals by observation of the position and height of the K conversion line of the 661-kev gamma-ray.

The region below 320 kev was studied with a Nylon window of thickness approximately 18 μg cm^{-2}, mounted on Lektromesh. The low energy region was rerun with a 12 μg cm^{-2} Nylon window. This window gave the same counting rate as the 18 μg cm^{-2} window down to 15 kev. This indicates that the thicker window transmits completely above 15 kev. A 1.2-mg cm^{-2} Dacron window was used to study the region above 100 kev. Between 170 and 300 kev the ratio of the counting rates with the two windows was constant. This ratio gives the relative transmission of the two windows. The energy calibration was based on the K conversion line of the 661-kev[30] gamma-ray of the cesium. In analysis of the Cs—Ba data a Fermi plot of

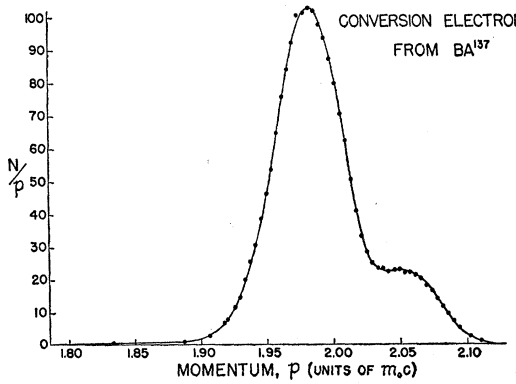

FIG. 3. K and L conversion lines of the 0.661-Mev gamma-ray of Cs^{137}.

[30] L. M. Langer and R. D. Moffat, Phys. Rev. 78, 74 (1950).

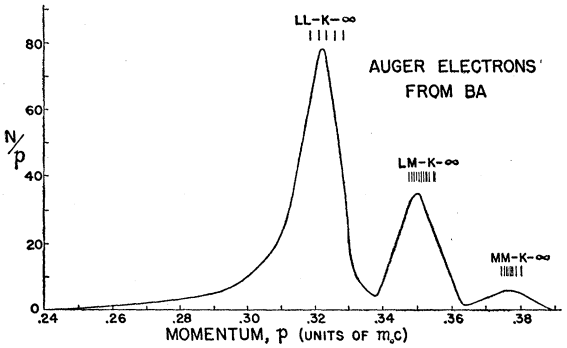

FIG. 4. Auger electrons of Ba from the Cs^{137} source.

the 1200-kev beta-group was made. This plot confirms the classification of this transition as second forbidden,[31] with spin change of 2, no parity change, and tensor interaction. By extrapolation of the straight line Fermi plot to low energy, this beta-group was subtracted from the 512-kev beta-group and the electron lines. Figure 2, a Fermi plot of the latter spectrum with the forbidden factor $C_{1T}=(W_0-W)^2+A(Z,W)(W^2-1)$,[32] gives a good straight line above 100 kev. The upward deviation below 100 kev is believed to be due to the inaccuracy for the forbidden factor[33] for small momenta and to the presence of the 90-kev beta-group of Cs^{134}. Since the Fermi plot was not a straight line in the region of the Auger electron lines, the continuous spectrum beneath these lines was obtained by a smooth joining of the continuous spectrum on each side of the lines.

Figures 3 and 4 show the conversion lines and Auger lines as counting-rate-per-unit-momentum interval *versus* momentum. The vertical lines above the Auger peaks mark the energy of the Auger electrons as calculated from x-ray spectra data given by Siegbahn.[34] For these calculations the energy levels of barium were used except for the level of the ejected electron. For this level, the value for the next higher atomic number element was used in order to approximate the doubly ionized state. The area beneath the curves is proportional to the intensity of the electrons producing the lines. The K to L–M conversion ratio of 4.64 measured by Graves *et al.*[35] was used to obtain the K conversion intensity from the measured total conversion intensity. The K Auger yield is the ratio of the K Auger electron intensity to the K conversion electron intensity. For barium this was found to be $a_K=0.130\pm0.007$. The ratios of the three K Auger lines were found to be K–LL:K–LX:K–XY::1.00:0.40±0.04:0.08±0.02. The probable errors given for the above values were obtained from a consideration of the statistical errors

[31] L. M. Langer and R. D. Moffat, Phys. Rev. 82, 635 (1951).
[32] J. P. Davison, Phys. Rev. 82, 48 (1951).
[33] M. E. Rose, of the Oak Ridge National Laboratory, is now calculating these factors accurately.
[34] M. Siegbahn, *Spektroskopie der Röentgenstrahlen* (Julius Springer, Berlin, 1931), p. 346.
[35] Graves, Langer, and Moffat, Phys. Rev. 88, 169 (1952).

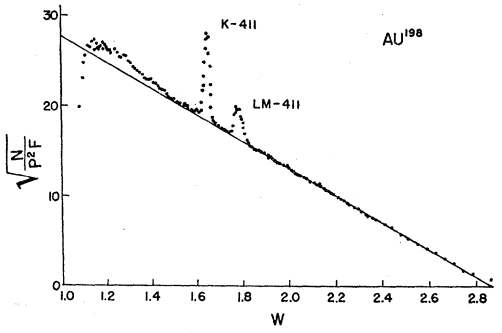

FIG. 5. Fermi-Kurie plot of the spectrum of the Au^{198} source.

of the individual points and what was believed to be the upper and lower limits for the position of the curves of the continuous spectrum beneath the lines.

B. Au^{198}—Hg^{198}

The data for the gold source were taken and analyzed in manner similar to that used for the cesium. The same 1.3 mg cm^{-2} Dacron window was used for the gold as for the cesium, but the thin window was made of collodion and was approximately 20 μg cm^{-2}. The K conversion line of the 411.2-kev gamma-ray[36] was used for calibration.

An allowed Fermi plot of the data taken with the thicker window is shown in Fig. 5. The deviation of the plot from linearity for energies below that of the K conversion line clearly shows the presence of low energy beta-groups. Figure 6 is a graph of the spectrum (taken with the thin window) in the region of the Auger lines after the 960-kev beta-group has been subtracted by extrapolation of the straight line Fermi plot. The lines marked A_1 and A_2 are the K Auger lines of mercury. The lines marked K_2, L_2, and M_2 are the K, L, and M conversion lines of the 159-kev gamma-ray, and K_3 is the K conversion line of the 209-kev gamma-ray of Au^{199}. The unmarked lines were unidentified. All attempts to identify these with known conversion lines of any impurity were unsuccessful. The presence of the Au^{199} conversion lines and their known intensities[37] were used to help fix the position of the continuous spectrum beneath the Auger electron lines. In the calculation of the K Auger yield, it was assumed that all of the K shell vacancies resulted from K conversion of the 159-, 209-, and 411-kev gamma-rays. The intensity of the K–XY Auger line relative to that of the K–LL line was assumed to be that calculated by Pincherle.[4] This resulted in a K Auger yield of $a_K=0.054\pm0.008$. The ratio of intensities of the two observed Auger lines was K–LL:K–LX::1.00:0.71±0.15. The percentage error is considerably greater for this measurement than for the barium Auger measurement. This is principally due to the fact that the continuous spectrum intensity is much larger in comparison with the Auger electron intensity than in the case of the barium measurements. If the unidentified lines were all K-conversion lines that contribute to the observed Auger electron peaks. the computed Auger yield would be reduced by three percent.

If K capture transitions occur in gold, they must give rise to platinum Auger electrons which would be approximately eight percent lower in energy than the mercury Auger electrons. It was estimated that if K capture transitions were 0.5 percent of the disintegrations, the presence of the platinum Auger electrons could have been detected.

C. Sn^{113}—In^{113}

In the work with the Sn^{113} source the region from 0.55 amp to 11.0 amp was covered by 400-count measurements in steps of 1 percent variation. Steps of 4 percent were used from 11.0 to 13.0 amp. On the peaks the number of counts per point was changed to 1000 and for the highest peaks to 3000, while the size of the current variations was reduced to about 0.3 percent. The region below the Auger peaks was covered four times; the Auger peaks nine times; the region between the Auger peaks and the conversion peaks twice; the conversion peaks eight times; the region above the conversion peaks twice. These data were taken with a four-layer (36-μg cm^{-2}) Nylon window. Figures 7 and 8 show the Auger peaks and conversion peaks, respectively. In order to guarantee that this window transmitted substantially all of the 20-kev Auger electrons, the Auger and conversion electron peaks were re-run with a window of only two layers (18 μg cm^{-2}) made at the same time as the four-layer window and with essentially identical layers. The 20-kev K–LL Auger peak was increased 3.8 percent by use of the thinner window. The cutoff of the thin window was below 3 kev, as is evidenced by the detection of a weak L Auger peak at 3.1 kev.

The 624-kev conversion line of Cs^{137} was used for energy calibration. Because of the poor resolution and a slight drift at the current necessary for the Cs^{137} line, the energies are not accurate to better than one percent. Within this error the K conversion line energy agrees with previous measurements, and the K Auger line

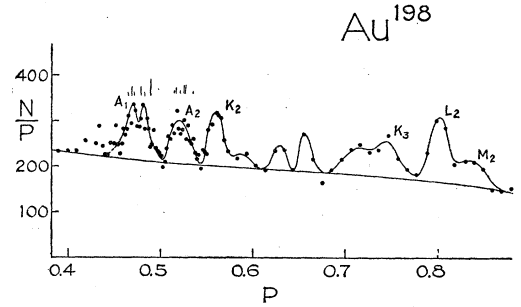

FIG. 6. The low energy portion of the spectrum of the Au^{198} source after subtraction of the 0.96-Mev spectrum of Au^{198}.

[36] DuMond, Lind, and Watson, Phys. Rev. **73**, 1392 (1948).
[37] P. M. Sherk and R. D. Hill, Phys. Rev. **83**, 1099 (1951).

energies agree with those calculated from x-ray measurements. A search was made for conversion lines of the 85-kev gamma-ray previously reported.[38] No conversion lines of intensity greater than one percent of the 360-kev line were found.

The experimental ratio of the K Auger intensity to the total conversion intensity (Figs. 7 and 8) is 0.54. In a previous report[39] a correction of 5 percent for window transmission was added. The work on $Cs^{137}-Ba^{137}$ reported above indicates that in all probability the 18 μg cm^{-2} window transmits all electrons of 20 kev. A 4 percent correction for transmission of the conversion lines through the Lektromesh gives a final figure of 0.56 ± 0.03 for the ratio of K Auger intensity to conversion line intensity.[40]

The ratios of K–LL to K–LX to K–XY Auger electrons can be very accurately determined from Fig. 7 because the lines are fairly well resolved, and there is no beta-ray background as in the case of Ba^{137} and Hg^{198}. The ratios are K–LL:K–LX:K–XY::1.00:0.417 $\pm$0.016:0.076$\pm$0.008.

V. INTERPRETATION AND SUMMARY

A. K Fluorescence Yield and K Auger Yield

In studying the available theory and experimental results on K fluorescence and K Auger yield, one finds[1,7] that for elements of low atomic number $w_K \ll 1$ and $a_K \approx 1$ and for elements of high atomic number $w_K \approx 1$ and $a_K \ll 1$. Thus, it would seem that direct K Auger yield measurements would be more accurate than K Auger yields obtained from fluorescence yield measurements for elements of atomic number greater than 40 since $a_K > 0.5$ for these elements. Measurements of w_K based on x-ray intensities greatly outnumber the measurements of a_K based on electron intensities, but

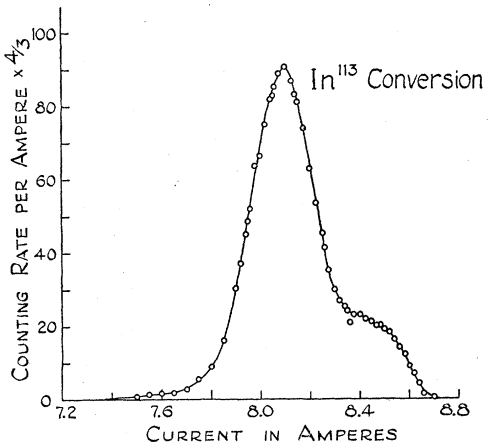

FIG. 7. K and L conversion lines of the 0.3917-Mev gamma-ray of In^{113}.

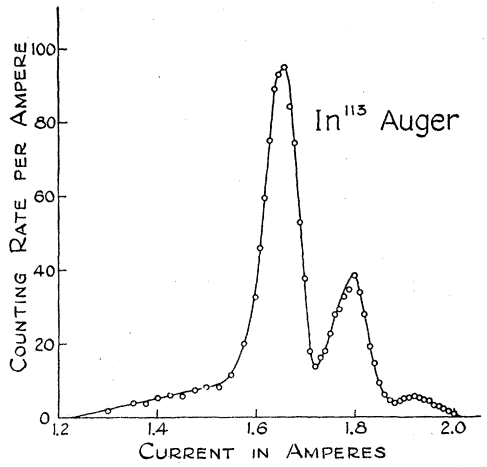

FIG. 8. Auger lines of In from the Sn^{113} source.

the former are confined principally to the elements with atomic number less than 50. The few cloud-chamber and photographic plate measurements of double electron tracks have been distributed over the whole atomic table and, in general, give values of w_K smaller than those obtained by other methods. Magnetic spectrometer measurements have been made only for elements with atomic number greater than 46.

In the present summary of the experimental results, the values are reported as $w_K = 1 - a_K$. Table II lists the numerical values followed by a lower case letter referring to the list of workers who obtained these values. This list gives the reference to the work and an indication of how the measurements were made. Figure 9 is a graph of these values. It appears that the $CZ^4/(a+Z^4)$ relation does not give a very good fit to these data, and that the proper analytical expression for the curve should probably involve an additional term or terms to include screening and relativistic effects. A least-squares fit to all these data would require a very thorough study of all the methods used, so that proper weights might be assigned the different values. The present authors have not attempted to carry out such a curve-fitting process, but have drawn an estimated curve that takes into consideration, in a rough way, what knowledge they have of the reliability of the various values. It is believed that the magnetic spectrometer measurements are the most accurate for the region in which they have been made.

In addition to the values of Auger yield, there are sufficient experimental measurements of the K–LX to K–LL Auger electron intensity ratios to indicate roughly the variation of this ratio with atomic number. Besides the ratios for indium, barium, and mercury reported in this paper, several measurements by other workers were mentioned in Sec. II. Figure 10 is a plot of these K–LX to K–LL ratios against atomic number. It is seen that this ratio increases with increasing atomic number, and for high atomic number is approximately that

[38] S. W. Barnes, Phys. Rev. **56**, 414 (1939).
[39] Thomas, Haynes, Broyles, and Thomas, Phys. Rev. **82**, 961 (1951).
[40] This figure replaces that previously reported in reference 39.

TABLE II. Summary of fluorescence yield measurements.

Element	Atomic number		K fluorescence yield							
C	6		0.0009[r]							
N	7		0.0015[r]							
O	8		0.0022[r]	0.082[j]						
Ne	10		0.0081[r]	0.083[j]						
Mg	12				0.013[l]					
Si	14				0.038[l]					
S	16				0.083[l]				0.15[n]	
Cl	17		0.04[u]		0.108[l]					
A	18		0.07[d]	0.149[j]	0.077[o]	0.06[v]	0.123[cc]			
Ca	20				0.15[l]				0.207[n]	
Cr	24		0.26[a]	0.26[b]	0.263[l]				0.265[n]	
Fe	26		0.37[a]		0.375[e]	0.314[f]	0.32[g]		0.343[n]	0.30[p]
Co	27							0.38[m]		
Ni	28		0.45[a]	0.385[t]	0.445[e]	0.374[i]	0.39[g]	0.39[m]	0.436[n]	0.364[p]
Cu	29		0.48[a]	0.41[t]	0.495[e]	0.419[f]	0.45[g]	0.43[m]	0.55[c]	0.401[p]
Zn	30		0.58[a]	0.48[t]	0.57[e]	0.444[f]	0.52[g]	0.45[m]	0.476[n]	0.450[p]
As	33							0.53[m]		
Se	34		0.68[h]	0.575[t]		0.578[f]	0.547[i]	0.55[m]	0.585[n]	0.550[p]
Br	35		0.68[h]	0.57[b]	0.56[k]		0.565[i]			
Kr	36		0.51[d]	0.53[s]	0.67[bb]					
Sr	38					0.644[f]		0.72[m]		
Zr	40			0.69[t]						
Mo	42		0.785[q]	0.735[t]	0.97[e]	0.826[f]	0.68[i]	0.79[m]		0.724[p]
Rh	45		0.801[q]	0.77[t]						
Pd	46		0.835[q]							
Ag	47		0.838[q]	0.81[t]	0.88[e]	0.83[ff]		0.72[m]	0.93[c]	0.795[p]
Cd	48		0.846[q]	0.79[t]		0.83[ff]		0.70[m]		
Sn	50		0.855[q]	0.81[t]				0.66[m]		0.825[p]
Sb	51		0.862[q]					0.64[m]		
Te	52		0.872[q]					0.59[m]		
I	53		0.88[h]							
Xe	54		0.70[d]	0.78[s]	0.81[bb]					
Cs	55		0.890[z]							
Ba	56		0.900[q]	0.870[gg]						
Pr	59		0.88[dd]							
Pt	78		0.942[x]							
Hg	80			0.946[gg]						
Pb	82	>	0.89[ee]							
Bi	83		0.934[v]	0.976[w]						
Po	84		0.894[aa]							

[a] W. Kossel, Z. Physik **19**, 333 (1923); x-ray intensity, ionization chamber, corrected for K jump by Broyles, Thomas, and Haynes.
[b] W. Bothe, Physik. Z. **26**, 40 (1925); x-ray intensity, ionization chamber, from the results of Barkla and Thomas, corrected for K jump by Broyles, Thomas, and Haynes.
[c] W. Bothe, Z. Physik **37**, 547 (1926); x-ray intensity.
[d] P. Auger, Ann. phys. **6**, 183 (1926); electrons, cloud chamber.
[e] M. Balderston, Phys. Rev. **27**, 676 (1926); x-ray intensity, ionization chamber, w_K as calculated by Backhurst.
[f] I. M. Harms, Ann. Physik **82**, 87 (1927); x-ray intensity, ionization chamber, w_K as calculated by Backhurst with minor corrections of Compton.
[g] L. H. Martin, Proc. Roy. Soc. (London) **A115**, 420 (1927); x-ray, ionization chamber, absorber.
[h] L. H. Martin, Proc. Roy. Soc. (London) **A115**, 420 (1927); deduced from Barkla and Philpot, Phil. Mag. **25**, 849 (1923) and Beatty, Proc. Roy. Soc. **A85**, 329 (1911), x-ray, ion chamber, change of ionization K edge.
[i] A. H. Compton, Phil. Mag. **8**, 961 (1929); x-ray intensity, ionization chamber.
[j] G. L. Locher, Phys. Rev. **40**, 484 (1932); electrons, cloud chamber, preferred values of Locher which seem also preferred in (1952).
[k] W. Stockmeyer, Ann. Physik **12**, 71 (1932); x-ray intensity, double ionization chamber, K edge.
[l] M. Haas, Ann. Physik **16**, 473 (1933); x-ray intensity, ionization chamber.
[m] D. K. Berkey, Phys. Rev. **45**, 437 (1934); x-ray intensity, ionization chamber.
[n] H. Lay, Z. Physik **91**, 533 (1934); x-ray intensity, photographic plate.
[o] Martin, Bower, and Laby, Proc. Roy. Soc. (London) **A148**, 40 (1935); electrons, cloud chamber.
[p] E. Arends, Ann. Physik **22**, 281 (1935); x-ray intensity, ionization chamber.
[q] I. Backhurst, Phil. Mag. **22**, 737 (1936); x-ray intensity, ionization chamber.
[r] W. Crone, Ann. Physik **27**, 405 (1936); x-ray intensity, ionization chamber, and counter.
[s] L. H. Martin and F. H. Eggleston, Proc. Roy. Soc. (London) **A158**, 46 (1936); electrons, cloud chamber.
[t] R. J. Stephenson, Phys. Rev. **51**, 637 (1937); x-ray intensity, ionization chamber.
[u] Weimer, Kurbatov, and Pool, Phys. Rev. **66**, 209 (1944); K-capture isotope, ionization chamber, window absorption.
[v] B. B. Kinsey, Can. J. Research **A26**, 421 (1948); data of Ellis, Proc. Roy. Soc. **A139**, 336 (1933); magnetic spectrum.
[w] B. B. Kinsey, Can. J. Research **A26**, 421 (1948); data of Flammersfeld, Z. Physik **114**, 227 (1939); magnetic spectrometer.
[x] Steffen, Huber, and Humbel, Helv. Phys. Acta **22**, 167 (1949); conversion electrons, magnetic spectrometer.
[y] Curran, Angus, and Cockroft (1949) value given by E. H. S. Burhop, *The Auger Effect and Other Radiationless Transitions* (Cambridge University Press, Cambridge, England, 1952), p. 28; proportional counter spectrometer.
[z] I. Bergstrom and S. Thulin, Phys. Rev. **79**, 539 (1950); conversion electrons, magnetic spectrometer.
[aa] L. S. Germain, Phys. Rev. **80**, 937 (1950); electrons, K-capture isotope, photographic plate.
[bb] D. West and D. Rothwell, Phil. Mag. **41**, 873 (1950); x-ray intensity, proportional counter spectrometer.
[cc] D. L. Dexter and W. W. Beeman, Phys. Rev. **81**, 456 (1951); width of x-ray absorption line.
[dd] E. Kondaiah, Phys. Rev. **83**, 471 (1951); conversion electrons, magnetic spectrometer.
[ee] H. M. Neuman, and I. Perlman, Phys. Rev. **81**, 958 (1951); deduced from Auger and conversion line intensities in magnetic spectrometer given by these authors without any allowance for K-capture transitions present. These transitions would probably increase the value to more than 0.95.
[ff] Huber, Humbel, Schneider, Shalit, Helv. Phys. Acta **25**, 3 (1952); coincidence measurements, magnetic spectrometer.
[gg] Broyles, Thomas, and Haynes, conversion electrons, magnetic spectrometer.

obtained from Pincherle's[4] Auger transition probabilities.

B. Discussion of Sn^{113}—In^{113}

The summary of fluorescence yields given above indicates that the Auger yield of indium is appreciably less than the 0.20 lower limit assumed in a preliminary report of the work on Sn^{113}.[40] The conclusions reached there concerning the energy of the orbital capture transition to the 391.7-kev[35] state of In^{113}, concerning the parity of Sn^{113}, and concerning the impossibility of another transition preceding the 391.7-kev transition, are therefore unjustified. Cork[41] has since reported weak conversion lines which were assigned to gamma-rays of 255.2 kev and 400.9 kev. The data reported in Sec. IV indicate that conversion lines of the former must have an intensity of less than 1 percent of the conversion lines of the 391.7-kev gamma-ray. Conversion lines of the latter if present would not have been resolved from those of the 391.7-kev gamma-ray. That the 400.9-kev conversion lines are extremely weak compared to the 391.7-kev lines is indicated by the failure of Cork *et al.*[41] to observe the L line of the former whereas the M line of the latter was detected. Absorption curves, in addition to indicating the absence of 85-kev[38] gamma-radiation, indicate that the 255-kev radiation, if present, is probably an order of magnitude weaker than the 391.7-kev radiation.[38,39] That the intensity of the 400.9-kev gamma-radiation cannot be more than about 5 to 10 percent of that of the 391.7-kev gamma-ray is indicated by the close check between the

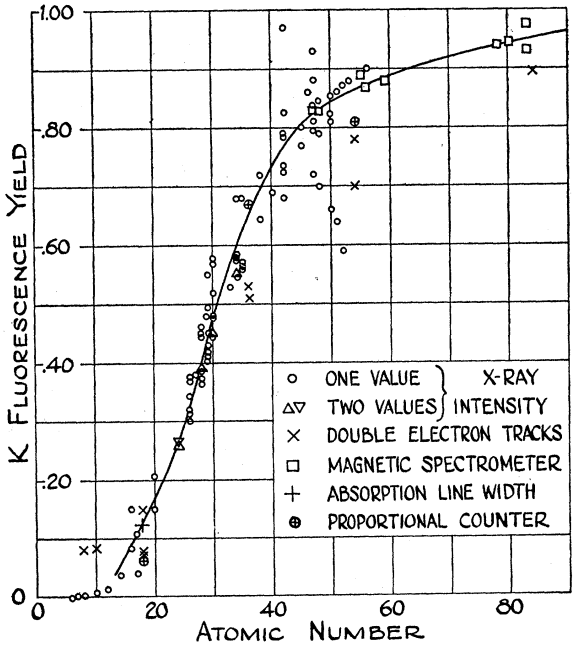

FIG. 9. Graphical summary of fluorescence yields.

[41] Cork, Stoddard, Branyan, Childs, Martin, and Leblanc, Phys. Rev. 84, 596 (1951).

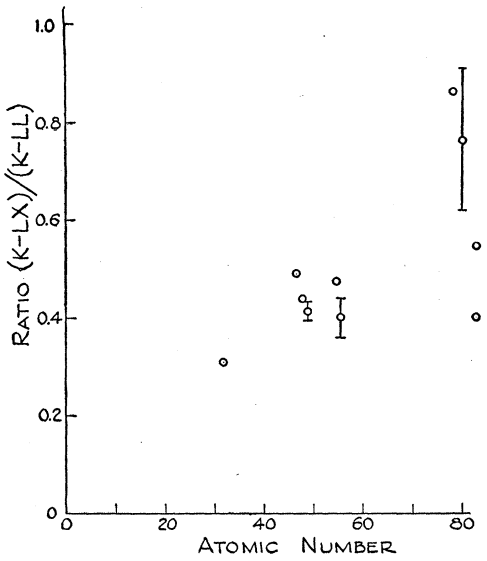

FIG. 10. Graphical summary of the ratio of K–LX to K–LL transitions as a function of atomic number as measured by various observers.

K conversion coefficient (0.445) of the latter gamma-ray obtained from the measurements of Cook and Haynes[42] and Graves, Langer, and Moffat[35] and the theoretical value for $M4$ radiation (0.47) computed by Rose *et al.*[43] Whether the weak transitions, if present, precede the 391.7-kev transition or are an alternative mode of disintegration is uncertain.

To illustrate the significance of the measured ratio of K Auger electrons to total conversion electrons, the decay scheme may be assumed to consist of a fraction p of the disintegrations through the 391.7-kev state and a fraction q of the disintegrations to the ground state through other levels but not through the 391.7-kev state. The ratio R of K Auger electrons to total conversion electrons is given by

$$R=R_p+a_K(q/p)(1+\alpha)/\alpha(1+\lambda_q), \tag{4a}$$

where

$$R_p=(a_K/\alpha)[(1+\alpha)/(1+\lambda_p)+\alpha_K], \tag{4b}$$

and λ_p and λ_q represent the ratio of L to K capture of the two branches, α and α_K represent, respectively, the conversion and K conversion coefficient of the 391.7-kev transition (other conversion lines have been neglected as negligible), and a_K represents the Auger yield of indium. R_p is the value if all transitions go through the 391.7-kev state.

The value of $\alpha_K=0.47\pm0.01$ is taken from Rose *et al.*[43] for $M4$ radiation. The value of $\alpha=0.58\pm0.01$ is computed from α_K with the help of the value of $\alpha_K/\alpha_{L+M}=4.21$ of Graves *et al.*[35] From the fluorescence yield curve $a_K=0.165\pm0.01$. With these values, the measured value of R, and by use of first-order error

[42] T.B. Cook, Jr., and S. K. Haynes, Phys. Rev. 86, 190 (1952).
[43] Rose, Goertzel, Spinrad, Harr, and Strong, Phys. Rev. 83, 79 (1951).

theory one obtains $\lambda_p=0.08\pm0.09$ for $R=R_p$, and $\lambda_p=0.18\pm0.09$ if 10 percent of the transitions go by an alternative route. The error is almost entirely due to the error in Auger yield. This fact indicates the great need for still more accurate knowledge of the value of a_K as a function of Z. It is clear, however, that if in 30 percent or more of the transitions of some radio-nuclide an L electron were captured ($\lambda_p>0.5$), the method outlined here would make possible a fairly accurate determination of the amount of L capture even with the present uncertainty in the value of a_K. An estimate of the disintegration energy could then be made by use of the results of Rose and Jackson[44] and Marshak[45] as previously indicated.[39]

[44] M. E. Rose and J. L. Jackson, Phys. Rev. **76**, 1540 (1949).
[45] R. E. Marshak, Phys. Rev. **61**, 446 (1942).

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Neutron Energy Distribution from the Proton Bombardment of Li, Be, and C at 375 Mev*

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High energy neutrons were produced by bombarding lithium, beryllium, and carbon targets with protons having a maximum energy of 375 Mev. The energy spectrum of the neutrons emitted in the forward direction was measured by determining the range spectrum of recoil protons scattered at 45° by a polyethylene scatterer. A telescope of stilbene crystals with copper range-determining absorbers was used as a detector. The neutron beam was monitored by a bismuth fission ionization chamber. For elements studied, the width of the neutron energy distribution broadens and the energy of the peak of the distribution decreases as the atomic number of the target material increases.

I. INTRODUCTION

AN investigation[1] was made of the energy spectra of neutrons emitted in the forward direction when several light elements were bombarded by 375-Mev protons. Targets of lithium, beryllium, and carbon were exposed to the internal proton beam of the Columbia University Nevis cyclotron. The energy of the protons was determined from the value of the magnetic field and the radial position of the target. The neutrons produced scattered protons in a polyethylene block. The range in copper of the scattered protons was determined, using a scintillation counter telescope. The energy distribution of the neutrons was then inferred from this proton-range spectrum.

II. DESCRIPTION OF THE EXPERIMENT

A diagram of the experimental arrangement is given in Fig. 1. The targets were placed inside the vacuum tank of the cyclotron in such a position that the neutrons emitted in the forward direction passed through collimating holes in the concrete shielding surrounding the cyclotron. The targets were rectangular slabs held by a clamp. All parts of the clamp were at least 2 in. from the leading edge of the target, so the clamp would not contribute to the neutron flux. The target thicknesses were 2 in. for lithium, $1\frac{3}{4}$ in. for beryllium, and $1\frac{1}{4}$ in. for carbon. The thicknesses of the beryllium and carbon targets were adjusted to give the same multiple scattering of protons, and hence to give roughly the same number of multiple traversals of the proton beam in the two targets. The targets were of sufficient height to intercept a major fraction of the internal beam of the cyclotron.

The neutron beam passed through a $\frac{3}{4}$ in. Plexiglas window and was collimated by a steel tube 4 in. in diameter and 6 in. long, which was imbedded in the lead and concrete shielding of the cyclotron. Two additional blocks of concrete were added to give better shielding to the counters. The neutrons were scattered alternately by carbon and polyethylene scatterers placed in the beam. The scatterers were 4 in. square, and had a thickness of 2.13 g/cm^2 for the carbon, and 2.38 g/cm^2 for the polyethylene. The thicknesses were

* This research was supported by the joint program of the Office of Naval Research and the Atomic Energy Commission.
[1] A preliminary report of this work was given by Goodell, Loar, and Durbin, Phys. Rev. **83**, 234 (1951).