Because of the small distance between atoms A and B, a multipole expansion of the interaction Hamiltonian cannot be used for the evaluation of second-order forces between these two atoms. However, this quantity is the same as the second-order interaction between an isolated pair of atoms (AB), and, therefore, does not occur in the expression for the three-body interaction. It was shown that in the evaluation of the deviation from additivity no essential error is introduced by taking the same value for the average excitation energies which occur in the matrix summations for W'' and W_0''.

The relative contribution of three-body forces may be written as a sum of two terms: a "direct" term $[\Delta^2/(1-\Delta^2)]$ and an "exchange" term. The direct term is always positive, thus enhancing the interaction as compared with an additive sum. The exchange term is negative for hydrogen; the result for helium contains an additional positive term of higher order in the exchange integrals. For many-electron atoms a result of the same character must be expected. This can easily be seen from a discussion of the quantity $(H'^2)_{00}$. The zero-order wave function is of the antisymmetric type; therefore, $(H'^2)_{00}$ may be written as a sum of terms of alternating sign depending on whether the order of the permutation is even or odd. Accordingly, we first obtain the direct terms with a positive sign in which there is no exchange of electrons between the initial and final states. The next term involves an exchange of one electron on atom A with one electron on atom B and has therefore a negative sign. For two helium atoms, there is a higher exchange term in which both electrons on atom A are interchanged with both electrons on atom B; this term is positive. For atoms with more than two electrons, there are additional exchange terms resulting in contributions of higher order in the exchange integrals.

As was mentioned in the introduction, this calculation was carried out with the specific purpose of applying the result to the problem of the crystal structure of the rare gases. Since the result of this effect is to weaken the attractive forces, it will contribute to the stability of the face-centered cubic lattice (see, e.g., reference 9). A more detailed analysis of this problem will be given in a forthcoming publication.

ACKNOWLEDGMENTS

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Auger Effect in the Heaviest Elements*

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The Auger effect in the heaviest elements has been investigated with magnetic beta-ray spectrometers. K-Auger yields for ytterbium, polonium, and uranium of 0.064 ± 0.01, 0.058 ± 0.005, and 0.033 ± 0.010, respectively, were obtained. The K-Auger electrons of ytterbium and polonium were resolved into two groups, $K-LL$ and $K-LX$, according as two or one L electrons were involved. The ratios of the intensities of the $K-LX$ electrons to the $K-LL$ electrons for ytterbium and polonium were found to be 0.64 ± 0.05 and 0.55 ± 0.03, respectively. A summary of measurements of K-fluorescence yields and K-Auger electron intensity ratios is given. A revised decay scheme for Np^{236} and a redetermination of the half-life of At^{211} (7.20 ± 0.05 hours) is included.

I. INTRODUCTION

IN the study of radioactive nuclei, it often becomes necessary to know the number of K-electron shell vacancies produced by the electron capture process as well as by internal conversion of gamma radiation. Reorganization in such an ionized atom with a vacancy in the K-electron shell can take place in either of two ways.[1] First, a transition may occur during which an electron from a higher energy level fills the vacancy, and the excess energy is emitted as the characteristic electromagnetic radiation (K x-rays) of the element. Secondly, "radiationless" reorganization may take place by the transference of the excess energy to an electron in a higher energy level and the subsequent ejection of this electron from the atom. These ejected electrons are known as K-Auger electrons, and the process of their radiationless emission is known as the Auger effect.

If the magnitude of this effect is known, a determination of the number of K-shell vacancies can be made by a count of either the K x-rays or the K-Auger electrons. The K-Auger coefficient is defined as the number of K-Auger electrons emitted per K-shell vacancy. A more widely used term is the K-fluorescence yield w_f, defined as the number of the K x-radiations

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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. 477 ff.

emitted per K-shell vacancy. It is obvious that the sum of the K-Auger coefficient and the K-fluorescence yield is unity.

Recently, a compilation of K-fluorescence yields became available[2] but this unfortunately contained no information on w_f above polonium ($Z=84$). Because of the rather extensive research now being conducted on the heaviest elements, data on the Auger effect in the translead region would be highly desirable. This study attempts to remove an inconsistency in the value of w_f for polonium[3] and extend our knowledge of the Auger effect to atomic numbers greater than 84.

II. EXPERIMENTAL RESULTS

A. Thulium-170–Ytterbium-170

The summary of K-fluorescence yields published recently by Broyles *et al.*[2] contains no information on the elements between praseodymium ($Z=59$) and platinum ($Z=78$). Data on the Auger effect in this region would seemingly be quite useful as an aid to the extrapolation of the Auger coefficient to the heaviest elements. Thulium-170 ($Z=70$) appeared to be an ideal isotope to study in this region.

Thulium-170 decays to Yb^{170} by the emission of two beta groups. A 968-kev (maximum energy) beta group in 76 percent abundance decays to the ground state of Yb^{170}, and a 884-kev (maximum energy) beta group (24 percent) decays to an excited level in Yb^{170} 84.4 kev above the ground state. This excited state decays to the ground state by the emission of electric quadrupole radiation of 84.4 kev.[4] Although Tm^{170} is a shielded nucleus and decay by electron capture must be considered, an upper limit of 0.2 percent for this mode of decay has been set by Jaffe.[5] Therefore, the only K-electron shell vacancies produced during the decay of Tm^{170} arise from the K-shell internal conversion of the 84.4-kev gamma ray.

Since the K-Auger coefficient is the ratio of the number of K-Auger electrons emitted to the total number of K-electron shell vacancies, the K-Auger coefficient of ytterbium can be readily determined if the intensities of the K-Auger electrons and the K-shell internal conversion electrons of the 84.4-kev gamma ray are known.

TABLE I. Electron lines from Tm^{170} beta decay.

Gamma energy (kev)	Electron energy (kev)	Conversion shell	Intensity (arbitrary units)
84.4	23.0	Yb K	100
	74.8	Yb L	314
	82.4	Yb M	72
	84.0	Yb N	9.6
(K x-rays)	40.3–43.5	$K-LL$	3.9
	48.6–51.8	$K-LX$	2.5

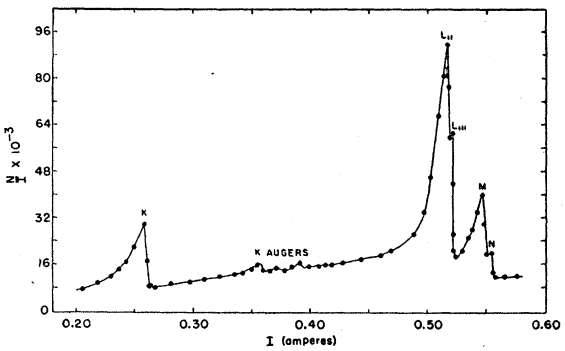

FIG. 1. Electron spectrum of Tm^{170}.

Thulium-170 was produced by the neutron irradiation of Tm^{169} (greater than 99 percent thulium) in the form of thulium oxide in the Materials Testing Reactor at the Reactor Testing Station, Arco, Idaho. Thulium-170, a 129-day negatron emitter, was purified by ion-exchange techniques using ammonium lactate as the eluant to separate the thulium from the other rare earths present as less than 1 percent impurity.

The electron spectrum was studied on the double-focusing beta spectrometer previously described by O'Kelley.[6] The detector has been changed to a side-window Geiger counter. The detector window consisted of three layers of a vinyl copolymer film supported by a grid of 0.001-inch tungsten wires spaced 0.008-inch apart on a copper ring. The window energy cutoff was about 4 kev and therefore the intensity corrections[7] are negligible. The experimental data are summarized in Table I. The electron spectrum is shown in Fig. 1 and the K-Auger electron spectrum is shown in greater detail in Fig. 2.

The K-Auger coefficient of ytterbium was found to be 0.064 ± 0.01 and the ratio of the groups of K-Auger lines to be $(K-LL):(K-LX)=1.00:(0.64\pm0.05)$ (where X denotes the M, N, etc., orbital-electron shells). The probable errors given were obtained from a consideration of the statistical errors of the individual points.

Table II compares the relative intensities of the internal conversion electron lines of the 84.4-kev gamma ray with those found by Graham *et al.*[4]

Graham *et al.*[4] did not resolve the N conversion electrons from the M conversion electrons, and therefore, their values for the ratios containing M should read $(M+N)$.

[2] Broyles, Thomas, and Haynes, Phys. Rev. **89**, 715 (1953).
[3] L. S. Germain, Phys. Rev. **80**, 937 (1950).
[4] Graham, Wolfson, and Bell, Can. J. Phys. **30**, 459 (1952); a better value for this energy is 84.26 ± 0.02 kev [E. N. Hatch and J. W. M. DuMond (private communication)].
[5] H. Jaffe, University of California Radiation Laboratory Report UCRL-2573, 1954 (unpublished).
[6] G. D. O'Kelley, University of California Radiation Laboratory Report UCRL-1243, 1951 (unpublished).
[7] R. O. Lane and D. J. Zaffarano, Phys. Rev. **94**, 960 (1954).

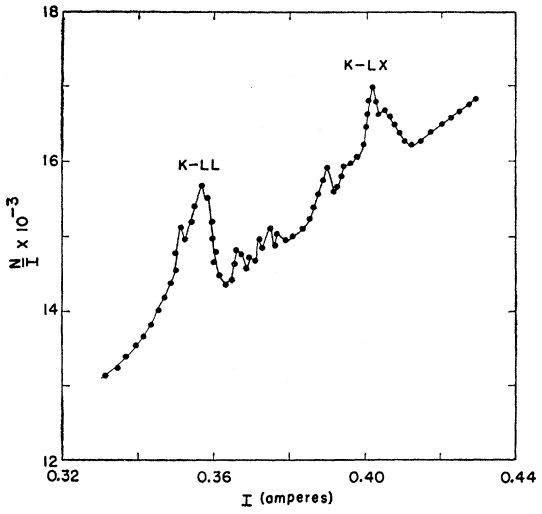

FIG. 2. K-Auger electron spectrum of Tm^{170}.

B. Astatine-211–Polonium-211

Astatine-211 was first made by Corson *et al.*[8] in bombardments of bismuth targets with 32-Mev helium ions. They suggested that At^{211} (7.3-hour half-life) showed branching decay, 60 percent electron capture to Po^{211}, and 40 percent alpha emission to Bi^{207}. The branching decay has subsequently been measured to be 59.1 percent electron capture and 40.9 percent alpha emission.[9] Polonium-211 decays by alpha emission with a half-life of 0.52 second[10] and is, therefore, in equilibrium with the At^{211}. The Bi^{207} daughter has a half-life which is long enough (27 years)[11] so that it may be neglected when studying At^{211}. A careful study has been made by Hoff[12] and Mihelich *et al.*[13] of the At^{211} electron-capture decay. No gamma radiation was found, with the possible exception of a (671±5)-kev gamma ray in very low abundance (0.37 percent of the astatine electron-capture decay). Although the assignment of this gamma ray to At^{211} is doubtful, its intensity is so low that its contribution to the Auger effect is negligible.

Germain[3] has studied the Auger effect in At^{211} by impregnating photographic emulsions with At^{211} and observing the alpha and Auger-electron tracks. His calculated value of 0.106 for the Auger coefficient of polonium appears to be large when compared with other values in the $Z=78$ to 83 region.[2,14]

The Auger effect of astatine can also be studied with the double-focusing beta spectrometer. Since every At^{211} electron capture is followed by an alpha emission from Po^{211}, the number of K-shell vacancies can be calculated by a determination of the absolute alpha disintegration rate. If the transmission of the spectrometer is known, the number of K-Auger electrons and subsequently the Auger coefficient of polonium can be calculated by an integration of the K-Auger electron spectrum.

The transmission of the double-focusing beta spectrometer was determined using a Th^{228} source. The experimentally integrated conversion-electron spectrum from the 84.3-kev gamma ray was compared with the known conversion-electron disintegration rate.

Astatine-211 was produced by the bombardment of bismuth metal which had been melted onto a 0.010-inch aluminum plate in a layer approximately 0.050 inch thick. These targets were clamped in a water-cooled target holder and mounted so as to intercept the full deflected helium-ion beam of the 60-inch cyclotron of the Crocker Radiation Laboratory of the University of California. Astatine-211, essentially free of other astatine isotopes, was produced by an $(\alpha,2n)$ reaction on the Bi^{209} target by attenuating the helium-ion beam to 29 Mev with 0.001-inch platinum foils.[15]

The requirement that the astatine be present in a narrow-line source necessitated modifications of the methods of Barton *et al.*[16] which utilize the high volatility of astatine in the zero valence state as compared to bismuth, lead, and polonium. It was found that temperatures as high as 600°C could be used without contaminating the astatine with bismuth, lead, or polonium. A line source was prepared for the double-focusing beta spectrometer by employing a 0.001-inch copper plate with a $\frac{3}{8}\times\frac{1}{16}$-inch slit as a collimating plate over a thin (157 micrograms/cm^2) palladium leaf. In later experiments, silver leaf was substituted for the palladium to utilize the greater affinity of silver for astatine.[17]

An alternate method was also employed for preparing samples of At^{211} for study in permanent-magnet beta spectrographs.[18] The bismuth target material was melted in a quartz tube in a vacuum system. A stream of nitrogen (1–2 mm pressure) prevented the astatine from condensing on the glass surfaces of the system.

TABLE II. Relative intensities of conversion electron lines of 84.4-kev gamma ray in the decay of Tm^{170}.

Ratio	Graham *et al.*[a]	This work
K/L	0.36±0.04	0.32±0.03
$K/(L+M)$	0.28±0.03	0.26±0.03
L/M	3.6 ±0.05	4.3 ±0.05

[a] See reference 4.

[8] Corson, MacKenzie, and Segrè, Phys. Rev. **57**, 459, 1087 (1944).
[9] H. M. Neumann and I. Perlman, Phys. Rev. **81**, 958 (1951).
[10] Leininger, Segrè, and Speiss, Phys. Rev. **82**, 334 (1951).
[11] G. Harbottle, quoted by D. E. Alburger and A. W. Sunyar, Phys. Rev. **99**, 695 (1955).
[12] R. W. Hoff, University of California Radiation Laboratory Report UCRL-2325, 1953 (unpublished).
[13] Mihelich, Schardt, and Segrè, Phys. Rev. **95**, 1508 (1954).
[14] M. Mladjenovic and H. Slätis, Arkiv Fysik **9**, No. 1, 41 (1955).
[15] E. L. Kelly and E. Segrè, Phys. Rev. **75**, 995 (1949).
[16] Barton, Ghiorso, and Perlman, Phys. Rev. **82**, 13 (1951).
[17] Johnson, Leininger, and Segrè, J. Chem. Phys. **17**, 1 (1949).
[18] A description of this instrument and its use will be published by W. G. Smith and J. M. Hollander from this laboratory.

The astatine was collected on a liquid-nitrogen cooled finger, coated with a thin layer of ice which contained a small amount of perchloric acid. The layer of ice was melted into a small centrifuge cone. Fifteen-mil silver wires, approximately $1\frac{1}{16}$ inches long, were placed in the melt, and the solution was stirred for approximately one hour. The astatine deposits on the wire in a manner thought to be analogous to the preparation of I^{131} silver-wire sources.[19] These wires were then mounted directly in the bent-crystal spectrometer, permanent-magnet beta spectrographs, and alpha-particle spectrograph.

Two separate and identical studies of the Auger effect in astatine were made using the double-focusing beta spectrometer. In the first study, a sample containing 7.2×10^8 disintegrations of At^{211} was sublimed onto palladium leaf as a source for the spectrometer. The K-Auger electron spectrum is shown in Fig. 3. While the resolution of the instrument does not allow the complete separation of the individual Auger lines, two distinct electron groups are clearly resolved. The first group of electrons, at an energy of 59 to 65 kev, was identified as the $K-LL$ Auger electrons. The second group of electrons at an energy of 72 to 76 kev was identified as the $K-LX$ electrons. The ratio of the two groups was found to be $(K-LL):(K-LX)::1.00:(0.55\pm0.03)$. The number of electrons in the K-Auger line spectrum was 4250. After correcting for the transmisson (0.022 percent) and the branching decay of At^{211} (59.1 percent electron capture), the Auger coefficient of polonium was calculated to be 0.049 ± 0.005. However, this value is calculated on the assumption of pure K-electron capture and must therefore be corrected for L-electron capture. The ratio of L-electron capture to K-electron capture should be about 0.18 according to Rose and Jackson[20] taking into account the decay energy.[21] Hoff,[12] using Germain's value of 0.106 for the Auger coefficient of polonium, experimentally determined the L- to K-electron capture ratio as approximately seven. Correcting this value using an Auger coefficient of 0.05 the experimental L- to K-electron capture ratio is six. After making this correction for L-electron capture in At^{211}, a value of 0.058 ± 0.005 as the Auger coefficient of polonium is obtained.

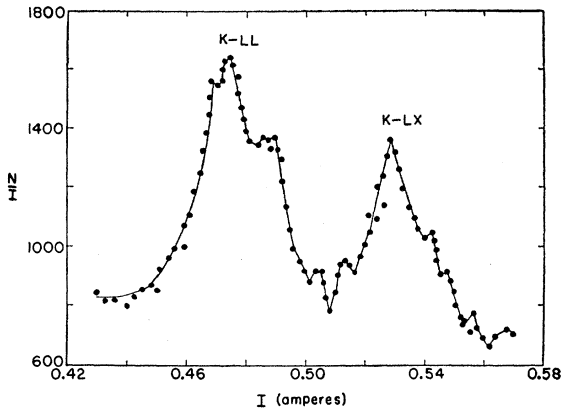

FIG. 3. K-Auger electron spectrum of At^{211}.

TABLE III. Energies and relative intensities of K-Auger electrons from At^{211} decay.

K-Auger electron	Energy experimental (kev)	Energy Bergström-Hill[a] (kev)	Relative intensity (arbitrary units)
$K-L_IL_I$	59.07	58.97	6.9
$K-L_{II}L_I$	59.75	59.67	10.0
$K-L_{III}L_I$	62.19	62.10	4.3
$K-L_{II}L_{III}$	62.83	62.80	9.4
$K-L_{III}L_{III}$	65.33	65.22	4.8
$K-L_{II}L_{II}$	...	60.37	<1
$K-L_IM_I$	72.01	71.98	...
$K-L_IM_{II}$	72.32	72.29	...
$K-L_IM_{III}$	72.91	72.86	...
$K-L_{II}M_{II}$		72.99	...
$K-L_IM_V$	73.62	73.47	...
$K-L_{II}M_{III}$		73.56	...
$K-L_{III}M_{II}$	75.46	75.41	...
$K-L_{III}M_{III}$	76.04	75.98	...

[a] See reference 21.

In the second determination of the Auger coefficient, At^{211} was sublimated onto palladium leaf to give a source containing 2.5×10^8 disintegrations per minute. The K-Auger electron spectrum obtained by using the double-focusing beta spectrometer contained 1425 electrons. After correcting for the transmission, the branching ratio of At^{211}, and the L-electron capture, a value of 0.056 ± 0.008 is obtained for the K-Auger coefficient of polonium which is in good agreement with the previous value. The ratio of the K-Auger electron groups was $(K-LL):(K-LX)::1.00:(0.61\pm0.08)$.

These values of the Auger coefficient of polonium are upper limits. Since the physical dimensions of the Th^{228} source from which the transmission was obtained was approximately twice the length of the At^{211} sources, this would give a lower limit to the transmission and hence an upper limit to the Auger coefficient.

This result of 0.058 for the Auger coefficient of polonium is in good agreement with the values expected in this region (see Sec. III).

Precise energies of the K-Auger electrons from At^{211} have been obtained by Smith and Hollander using their 50-gauss permanent-magnet spectrograph.[18] Table III lists the K-Auger electrons observed with their experimental energies.[22] Energies calculated using the Berg-

[19] W. G. Smith, University of California Radiation Laboratory Report UCRL-2974, 1955 (unpublished).
[20] M. E. Rose and J. L. Jackson, Phys. Rev. **76**, 1540 (1949).
[21] Glass, Thompson, and Seaborg, J. Inorg. Nuc. Chem. **1**, 3 (1955).

[22] Besides the K-Auger electrons, 46.0 ± 0.3-kev electrons were observed in varying intensities relative to the K-Auger electrons on several photographic films from the permanent-magnet spectrograph studies. While the varying relative intensity of these 46-kev electrons is very puzzling, it is believed that they are not due to impurities in the sample because weak 46-kev electrons have been observed in the electron spectrum of Em^{211} which decays to At^{211} by electron capture. These 46-kev electrons are

ström-Hill formula[23] and the x-ray absorption-edge energies of Hill *et al.*[24] are included for comparison. Experimental relative intensities of the $K-LL$ electrons are included. Intensities of the $K-LX$ electrons were too weak for measurement.

The half-life of At^{211} has also been redetermined. Corson *et al.*[8] and Kelley and Segrè[15] have reported the half-life of At^{211} to be 7.5 hours. Hall and Templeton[25] have reported 7.3 hours for the half-life and Neumann and Perlman[9] report 7.2 hours. Especially purified bismuth was bombarded with 29-Mev helium ions in the 60-inch cyclotron of the Crocker Radiation Laboratory. The radioactive alpha decay of an At^{211} sample mounted on silver to minimize loss of astatine by evaporation was followed for 12 half-lives in an argon-filled ionization chamber (52 percent counting efficiency). The radioactive decay curve of At^{211} was straight over the 12 half-lives. This experimentally determined half-life of At^{211} is 7.20 ± 0.05 hour.

C. Neptunium-236–Uranium-236

Since no experimental results are available on the Auger effect in the elements above polonium ($Z=84$), it would be of interest to measure the Auger coefficient of some nuclide in this region. Neptunium-236 had been studied previously by Orth and O'Kelley[26] who reported an electron capture to negatron emission ratio of two. Two negatron groups of maximum energy, 0.51 and 0.36 Mev, respectively, were detected as were x-rays and a gamma ray of 150 kev which was reported to be approximately 100 percent internally converted. No other gamma radiations were observed. This isotope, then, appeared to be ideal for studying the Auger effect since the number of K vacancies could be determined through a knowledge of the decay scheme, the transmission of the double-focusing beta spectrometer, and integrations of the K internal-conversion electron spectrum of the 150-kev gamma ray and the negatron spectrum. The number of K-Auger electrons and, hence, the Auger coefficient could then be determined from an integration of the K-Auger electron spectrum.

A source of Np^{236} was studied on the double-focusing beta spectrometer by T. O. Passell and the author. An electron spectrum was observed which was similar to that obtained in the previous work.[26] However, certain lines in the spectrum showed greater complexity than before. Coupled with recent gamma-ray and x-ray spectroscopy studies,[5] a reinterpretation of the data became necessary, and a new decay scheme was proposed.

The electron lines designated as L-Auger electrons by Orth and O'Kelley[26] were resolved into four lines which were interpreted as the L_{II} and L_{III} internal conversion electrons of gamma rays with energies 43.5 ± 1 kev and 44.2 ± 1 kev. A line previously reported as the K-conversion electron line of a 150-kev gamma ray may also be interpreted as the unresolved M-conversion electrons of the 43.5- and 44.2-kev gamma rays. Unresolved N-conversion electrons were also observed as were K-Auger electrons. The electron data are summarized in Table IV.

The assignment of the 43.5-kev gamma ray to Pu^{236} and the 44.2-kev gamma ray to U^{236} is quite arbitrary. The latter is in good agreement with the energy of the 44-kev determined[27] for the first excited state of U^{236} from the alpha-particle fine structure of Pu^{240}.

Since the intensity of the negatron spectrum observed on the double-focusing spectrometer was too low for a Fermi analysis, advantage was taken of the higher transmission of the magnetic-lens spectrometer to obtain these data. The Fermi-Kurie plot exhibited a marked concavity toward the abscissa which is indicative of a forbidden transition. The maximum energy obtained was 518 ± 10 kev.[28]

X-ray and gamma-ray spectroscopy revealed no evidence of a 150-kev gamma ray in a thorough examination of the 0- to 200-kev energy region.[5]

The K-electron-capture/negatron-emission ratio was determined by observing the alpha growth of the Pu^{236} daughter.[5] Alpha-pulse analysis indicated that only the 5.75-Mev alpha particles of Pu^{236} were present. The alpha-particle disintegration rate after the 22-hour Np^{236} had completely decayed, coupled with the

TABLE IV. Electron lines of Np^{236}.

Gamma energy (kev)	Electron energy (kev)	Conversion shell	Intensity (arbitrary units)
43.5 ± 1	21.1	Pu L_{II}	
	25.4	Pu L_{III}	48
44.2 ± 1	23.2	U L_{II}	
	27.0	U L_{III}	
$44\ \pm2$	37.9	Pu ΣM and U ΣM	18
$43\ \pm1$	41.7	Pu ΣN and U ΣN	3
K x-rays	88 ± 5	U ΣL and U ΣM	5
	(K-Auger electrons)		
...	500 ± 30 (β^- maximum)	...	200

not due to At^{210} contamination. The ratio of At^{210} to At^{211} produced has been measured to be less than 10^{-6} by determining the intensity of the 138-day alpha emitter Po^{210}, after all the At^{210} and At^{211} have decayed. A sample of At^{211} was also studied on a 10-inch bent-crystal photon spectrometer of the Cauchois type. Besides the K x-rays, electromagnetic radiation of 62.35 ± 0.5 kev were observed in low abundance (4 percent of the K x-rays). While definite assignments of the 46.0-kev electrons and the 62.3-kev electromagnetic radiation cannot be made, it may be that these electrons were also observed by Germain and misinterpreted as K-Auger electrons, giving an abnormally high value to his K-Auger coefficient.

[23] I. Bergström and R. D. Hill, Arkiv Fysik **8**, 21 (1954).
[24] Hill, Church, and Mihelich, Rev. Sci. Instr. **23**, 523 (1952).
[25] K. L. Hall and D. H. Templeton (unpublished, 1951).
[26] D. A. Orth and G. D. O'Kelley, Phys. Rev. **82**, 758 (1951).
[27] F. Asaro and I. Perlman, Phys. Rev. **88**, 828 (1952).
[28] G. D. O'Kelley (private communication, 1954).

absolute abundance of the K x-rays at the beginning of the experiment, indicated the K electron-capture/negatron-emission branching ratio of Np^{236} as (43±5 percent)/(57±5 percent). The K x-ray abundance was corrected for the presence of Np^{234} in the sample and a fluorescence yield of 0.97.

X-ray spectroscopy[5] indicated that the intensity of the L x-rays was 0.6 relative to the K x-rays. Making corrections for (1) a mean L-x-ray fluorescence yield of[29] 0.5, (2) a K x-ray fluorescence yield of 0.97, (3) 72 percent of the K-shell vacancies being filled by L-shell electrons,[30] and (4) 43 percent of the disintegrations being due to K-electron capture, there are a maximum of 0.48 L-shell vacancies from internal conversion per K electron-capture decay or 0.21 vacancy per disintegration.

From the electron intensities, the abundance of conversion electrons is about 35 percent of the negatron spectrum. Since 57 percent of the disintegrations take place by negatron emission, there are 20 conversion electrons per 100 disintegrations. Thus, within the limits of error of the intensity measurements, all of the L-shell vacancies can be accounted for by the L-shell internal conversion of the 43.5- and 44.2-kev gamma rays. However, since the limits of error are such that five to ten L-shell vacancies per 100 disintegrations could be arising from L-electron capture, it appears safe to conclude that the lower limit for the K/L-capture ratio is around four.

Since the intensities of the L electrons from the conversion of the 43.5- and 44.2-kev gamma rays are approximately equal, an upper limit of about ten events per 100 disintegrations populate each of the first-excited states of Pu^{236} and U^{236}.

A decay scheme, consistent with the available data, is given in Fig. 4. The log ft values for negatron decay to the ground state and excited state are 6.6 and 7.1, respectively, and the corresponding values for the electron capture decay are 7.0 and 7.5, respectively (0.9-Mev total decay energy[21]).

The equal intensities of the L_{II} and L_{III} conversion electrons suggest that both gamma rays are $E2$ transitions. This is consistent with the usual assignment in this region for even-even nuclides of zero spin and even parity for the ground state and spin 2 with even parity for the first-excited state.

A value of the K-Auger coefficient of uranium can now be obtained through a knowledge of the decay scheme and the relative intensities of the electrons. A knowledge of the transmission of the spectrometer is not necessary in this calculation. The relative intensity of the internal conversion electrons from both the 43.5- and 44.2-kev gamma rays is 69. Since each excited state in U^{236} and Pu^{236} is approximately evenly populated (20 percent) and the K electron-capture branching is 43 percent, we have a relative intensity of 30 for

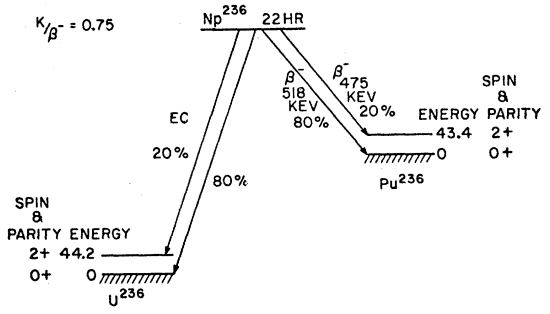

FIG. 4. Decay scheme of Np^{236}.

the 43.5-kev gamma ray resulting from the electron-capture decay of Np^{236}. Since these L internal conversion electrons occur in only 20 percent of the transitions, however, this corresponds to a total of 150 transitions. Assuming K-electron capture only, an assumption upon which the branching ratio was determined, this corresponds to 150 K-shell vacancies.

Because there are five K-Auger electrons observed relative to the 150 K-shell vacancies, the K-Auger coefficient of uranium is 0.033±0.01. The limit of error results from the uncertainty in the intensities of the electrons and in the K/L-electron-capture ratio.

III. INTERPRETATION AND SUMMARY

The first attempt to outline a theory of the Auger effect was by Wentzel[31] in 1927. Using a two-electron atom of high nuclear charge, Wentzel was able to show that the reciprocal of the lifetime of a K-shell vacancy with respect to an Auger transition in which an L electron is emitted should be independent of Z. Since the reciprocal of the effective life of an excited K state with respect to radiation of x-rays is approximately proportional[1] to Z^4, w_K, the K-fluorescence yield, may be expressed as

$$w_K = Z^4/(k+Z^4).$$

Such an expression, however, can only be qualitatively correct because 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[32] rather than only two. Such a theory in a statisfactory state does not exist at present.

By using the expression derived by Wentzel,[31] calculations of the fluorescence yield have been made by Burhop[33] and Pincherle[34] using a nonrelativistic theory and by Massey and Burhop[35] using a relativistic theory. In these calculations, hydrogen-like, single-electron wave functions were used, the effective nuclear

[29] B. B. Kinsey, Can. J. Research **26A**, 404 (1948).
[30] A. H. Compton and S. K. Allison, reference 1, p. 640.
[31] G. Wentzel, Z. Physik **43**, 524 (1927).
[32] N. F. Mott and I. N. Sneddon, *Wave Mechanics and Its Applications* (Oxford University Press, London, 1948), p. 338 ff.
[33] E. H. S. Burhop, Proc. Roy. Soc. (London) **A148**, 272 (1935).
[34] L. Pincherle, Nuovo cimento **12**, 81 (1935).
[35] H. W. S. Massey and E. H. S. Burhop, Proc. Roy. Soc. (London) **A153**, 661 (1936).

TABLE V. Comparison of the experimental and theoretical values of the Auger yield of ytterbium, polonium, and uranium.

Element	Atomic number	Auger yield experimental	theoretical[a]
ytterbium	70	0.06	0.08
polonium	84	0.06 (upper limit)	0.045
uranium	92	0.03	0.04

[a] See reference 35.

charge being determined by the application of Slater's rules.[36] The use of screened, hydrogen-like wave functions in the relativistic calculations increases the calculated Auger probabilitity for elements of high atomic number. The calculations involved are very laborious and have been carried out only for a few cases. The relativistic value of the K-Auger yield for mercury, for example, is 0.06 while the nonrelativistic value is only one-half as great, or 0.03. Relativistic effects are of some importance for elements of atomic number as low as 47 where the relativistic value of the Auger yield is still approximately 20 percent larger than the nonrelativistic value.

Table V compares the experimental values of the Auger yield of ytterbium, polonium, and uranium with the calculated theoretical values of Massey and Burhop.[35] The experimental results are only in qualitative agreement with the theoretical values as was expected. Until a more accurate theory of the Auger effect becomes available, quantitative agreement will probably not be possible.

Several summaries of the magnitude of the Auger effect have been published. The summaries of Arends[37] in 1935 and Backhurst[38] in 1936, for example, cover the contemporary data on fluorescence yields very well but do not include any measurements on the heavier elements. Not until the complete summaries of Burhop[39] (data through 1950) and Broyles *et al.*[2] became available did the first collected data on elements of atomic number greater than 56 appear. A new and, it is hoped, complete summary of the data through June, 1955 is given in Fig. 5. As the only new data are for elements with atomic numbers 40 and larger, a complete reference list is not included because Broyles *et al.*[2] have covered the reference material of their available data very thoroughly. The new values of the fluorescence yields for elements heavier than zirconium ($Z=40$) are listed in Table VI. The value is followed by a lower-case letter referring to a list of workers who obtained these values and an indication of how the measurements were made.

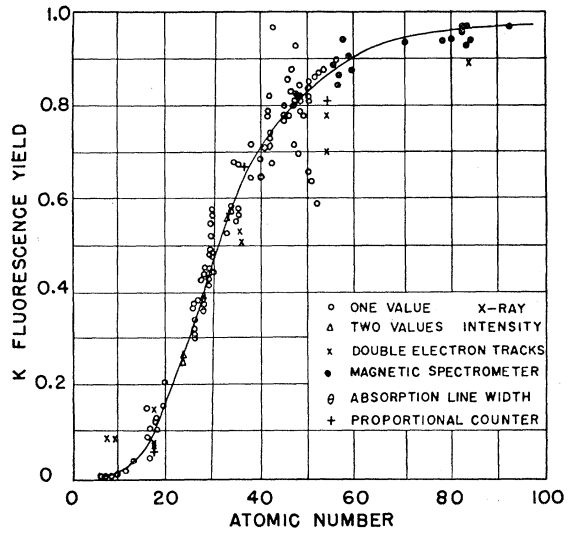

FIG. 5. Graphical summary of K-fluorescence yields.

TABLE VI. Recent data on fluorescence-yield values.

Element	Atomic number	K-fluorescence yield	Reference
zirconium	40	0.645	a
niobium	41	0.713	a
molybdenum	42	0.714	a
rhodium	45	0.779	a
palladium	46	0.781	a
silver	47	0.813	a
cadmium	48	0.819	a
tin	50	0.840	a
barium	56	0.85	b
lanthanum	57	0.94	c
cerium-praseodymium	58–59	0.90	d
ytterbium	70	0.94	e
lead	82	0.96	f
bismuth	83	0.96	g
polonium	84	0.94	h
uranium	92	0.97	h

[a] C. E. Ross, Phys. Rev. **93**, 41 (1954).
[b] T. Azuma, J. Phys. Soc. Japan **9**, 443 (1954), conversion electrons; magnetic spectrometer.
[c] C. H. Pruett and R. G. Wilkinson, Phys. Rev. **96**, 1340 (1954); conversion electrons, electron-capture isotope, magnetic spectrometer.
[d] Browne, Rasmussen, Surls, and Martin, Phys. Rev. **85**, 146 (1952); electron-capture isotope, magnetic spectrometer.
[e] This work, conversion electrons, magnetic spectrometer.
[f] A. H. Wapstra, Ph.D. thesis, University of Amsterdam (published by G. van Soest, Amsterdam, 1953); electron-capture isotope, conversion electrons, magnetic spectrometer.
[g] M. Mladjenovic and H. Slätis, Arkiv Fysik **9**, 41 (1955); conversion electrons, magnetic spectrometer.
[h] This work, electron-capture isotope, magnetic spectrometer.

A least-squares fit to all these data has not been attempted here, but an estimated curve has been drawn through the points taking into consideration the estimated reliability of the various values. Below $Z=40$, the curve is essentially the same as that presented by Broyles *et al.*,[2] and their analyses of the reliability of the values will suffice here also. Above $Z=40$, the curve varies slightly from Broyles *et al.*,[2] because of the additional data on this heavy-element region which has become available.

The experimental values support in a qualitative way the theoretical variation of the fluorescence yield with atomic number, but none of the curves yet

[36] J. C. Slater, Phys. Rev. **36**, 57 (1930).
[37] E. Arends, Ann. Physik **22**, 281 (1935).
[38] I. Backhurst, Phil. Mag. **22**, 737 (1936).
[39] E. H. S. Burhop, *The Auger Effect and Other Radiationless Transitions* (Cambridge University Press, Cambridge, 1952), pp. 44–51.

suggested, theoretical or semiempirical, is in satisfactory agreement with all the data. The relation

$$w_K=0.957Z^4(0.984\times10^6+Z^4)^{-1},$$

given by Arends[37] and recommended by Tellez-Plascencia[40] and Broyles *et al.*,[2] gives the best fit of those that have been proposed. If an expression of the form

$$w_K=AZ^4(b+Z^4)^{-1}$$

is a valid representation of the data, a plot of $1/w_K$ *versus* $1/Z^4$ should be linear with a slope of b/a. As shown in Fig. 6, the experimental results are well represented by a straight line drawn through the most reliable of the experimental values. The values of a and b which best represent the experimental points are 0.98 and 0.98×10^6, respectively. The expression of the K-fluorescence yield as a function of Z then becomes $w_K=0.98Z^4(0.98\times10^6+Z^4)^{-1}$.

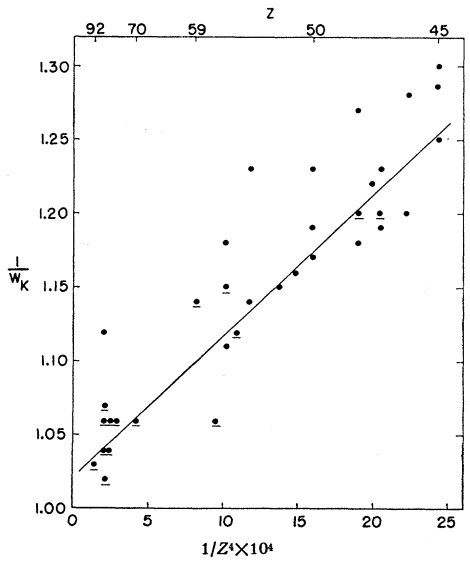

FIG. 6. Variation of $1/w_K$ with $1/Z^4$ in the heavier elements.

Since the available theory and experimental results on K-fluorescence and K-Auger yields agree that for elements of low atomic number $w_K\ll1$ and $a_K\approx1$ and for elements of high atomic number $w_K\approx1$ and $a_K\ll1$, it would appear that the direct measurement of the K-Auger yield would be more accurate than a direct determination of the fluorescence yield for elements of atomic number greater than 30 since $a_K>0.5$ for these elements. In general, this has been done experimentally and exclusively for elements of atomic number greater than 56. It is believed that the magnetic spectrometer measurements are the most accurate for the region in which they have been made. These data are indicated by underlining the experimental points in Fig. 6.

In addition to the values of the K-fluorescence yields

[40] H. Tellez-Plascencia, J. phys. radium **10**, 14 (1949).

TABLE VII. Experimental ratios of $K-LX$ to $K-LL$ transitions as measured by various observers.

Element	Atomic number	$(K-LX)/(K-LL)$ ratio	Reference
nickel	28	0.28	a
copper	29	0.292	b
copper	29	0.252	c
germanium	32	0.31	d
arsenic	33	0.25	e
bromine	35	0.33	f
strontium	38	0.28	g
silver	47	0.49	h
silver	47	0.63	i
cadmium	48	0.44	h
indium	49	0.42	j
cesium	55	0.48	k
barium	56	0.4	j
barium	56	0.5	l
ytterbium	70	0.64	m
platinum	78	0.56	n
mercury	80	0.71	j
mercury-thallium	80–81	0.53	o
thallium	81	0.55	p
bismuth	83	>0.4	q
bismuth	83	>0.57	r
bismuth	83	0.64	s
polonium	84	0.4	t
polonium	84	0.55	m
Theoretical value for all atomic numbers		0.716	u

[a] E. Plassman and F. R. Scott, Phys. Rev. **84**, 156 (1951).
[b] J. F. Perkins and S. K. Haynes, Phys. Rev. **92**, 687 (1953).
[c] T. Yuasa, Physica **18**, 1267 (1952).
[d] M. Ference, Phys. Rev. **51**, 727 (1937).
[e] F. R. Scott, Phys. Rev. **84**, 659 (1951).
[f] S. Thulin (private communication to I. Bergström), as reported in K. Siegbahn, *Beta- and Gamma-Ray Spectroscopy* (Interscience Publishers, Inc., New York, 1955), p. 633.
[g] J. Schooley (private communication).
[h] Huber, Humbel, Schneider, and de Shalit, Helv. Phys. Acta **25**, 3 (1952).
[i] T. A. Johnson and J. S. Foster, Can. J. Phys. **31**, 464 (1953).
[j] Broyles, Thomas, and Haynes, Phys. Rev. **89**, 715 (1953).
[k] I. Bergström and S. Thulin, Phys. Rev. **79**, 539 (1950).
[l] T. Azuma, J. Phys. Soc. Japan **9**, 443 (1954).
[m] Experimental work of author.
[n] Steffens, Huber, and Humbel, Helv. Phys. Acta **22**, 167 (1949).
[o] Passell, Michel, and Bergström, Phys. Rev. **95**, 999 (1954).
[p] C. J. Herrlander, Nobel Institute of Physics (private communication to I. Bergström), as reported in K. Siegbahn, *Beta- and Gamma-Ray Spectroscopy* (Interscience Publishers, Inc., New York, 1955), p. 633.
[q] B. B. Kinsey, Can. J. Research **A26**, 421 (1948), data of C. D. Ellis, Proc. Roy. Soc. (London) **A139**, 336 (1936).
[r] B. B. Kinsey, Can. J. Research **A26**, 421 (1948), data of A. Flammersfeld, Z. Physik **114**, 227 (1939).
[s] M. Mladjenovic and H. Slätis, Arkiv Fysik **9**, No. 1, 41 (1955).
[t] R. W. Hoff, University of California Radiation Laboratory Report UCRL-2325, September 1953 (unpublished).
[u] L. Pincherle, Nuovo cimento **12**, 81 (1935).

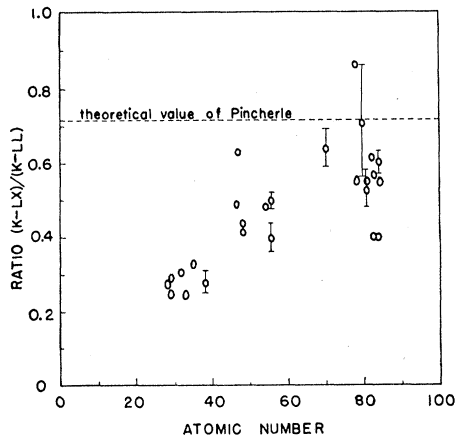

FIG. 7. Ratio of $(K-LX)/(K-LL)$ Auger electrons.

TABLE VIII. Experimental and theoretical relative intensities of K-Auger lines.

Element	Z	$K-L_IL_I$	$K-L_IL_{II}$	$K-L_IL_{III}$	$K-L_{II}L_{III}$	$K-L_{III}L_{III}$	$K-L_{II}L_{II}$	Reference
				K-Auger lines				
silver	47	1.0	1.3	1.3	3.2	1.8	0.5	a
xenon	54	weak	moderate	moderate	strong	moderate	...	b
gold	79	1.0	1.7	1.2	1.4	0.8	0.3	c
mercury	80	1.0	1.2	0.7	1.4	0.6	0.2	d
bismuth	83	1.0	1.8	1.1	1.6	0.8	<0.2	e
bismuth	83	1.0	1.8	1.3	2.3	1.3	<0.2	f
polonium	84	1.0	1.4	0.6	1.3	0.7	<0.1	g
theoretical nonrelativistic		1.0	1.13	2.26	4.03	2.30	0.38	h
theoretical relativistic		1.0	5.5	5.3	...	...	...	i

[a] See reference 41.
[b] See reference 19.
[c] J. W. Mihelich, Phys. Rev. **88**, 415 (1952).
[d] See reference 23.
[e] See reference 14.
[f] C. D. Ellis, Proc. Roy. Soc. (London) **A138**, 318 (1932); **A143**, 350 (1934).
[g] Experimental results of author.
[h] See reference 39.
[i] See reference 35.

there are sufficient experimental measurements of the $K-LX$ to $K-LL$ Auger electron intensity ratios available to indicate approximately the variation of this ratio with atomic number. The available data on the Auger electron intensity ratios are listed in Table VII and shown graphically in Fig. 7. It is interesting to compare these experimental ratios of the intensities of the groups of Auger electrons to those obtained from the calculated probabilities of Pincherle[34] using the expressions derived by Wentzel.[31] This calculated intensity ratio is $(K-LL):(K-LX)=1.00:0.716$ and should be independent of atomic number. As seen from Fig. 7, the experimental ratios are not constant, but appear to increase with increasing atomic number. For the heavier elements, the ratios approach the value obtained by Pincherle using transition probabilities.

It may be of interest also to compare the values of the relative intensities of the individual $K-LL$ Auger electrons of polonium with the other available data as well as with the theoretical relativistic intensities of Massey and Burhop[35] and the nonrelativistic calculations of Burhop.[33] This comparison is given in Table VIII.

The experimentally determined relative intensities of polonium are not in agreement with the theoretical predictions but agree fairly well with the other heavy-element values. The values calculated by Burhop[33] are for $Z=47$ but should be almost independent of Z in the nonrelativistic theory. It is apparent, however, that the experimental results of Johnson and Foster[41] for $Z=47$ are in somewhat better agreement with these nonrelativistic intensities than are the values of the heavier elements.

IV. ACKNOWLEDGMENTS

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[41] T. A. Johnson and J. S. Foster, Can. J. Phys. **31**, 464 (1953).