mine the relationship between the longitudinal and transverse fields, for there enters here the ratio of $p_L{}^M$ to $p_L{}'^M$ which requires a detailed knowledge of the charge-current distribution. The exceptional case is the long wavelength limit; in this case we have $p_L{}^M = p_L{}'^M$ and then the condition that the current be bounded establishes a unique connection between the longitudinal field and the transverse (electric type) field, and thus enables us to express the electric radiation in terms of the charge density.

Examining this from a slightly different point of view, it follows from (10) that for $kR \ll 1$ the most singular part of **E** is simply the longitudinal field. Thus in the long-wave limit, the electric field near the surface is purely longitudinal. In this limit, then, we can regard the electric type field as being determined by the wave equation and the boundary condition that for small R its electric field reduce to the longitudinal field. Thus there arises the connection between the two fields. Incidentally, we remark that in some calculations where the long-wavelength approximation is used from the beginning one may, because of the close connection between the fields, encounter an ambiguity in separating the contributions of the longitudinal and transverse electric fields to the process in question. Such ambiguities are, of course, resolved by examining the transition from the rigorous solution to the long-wave limit.

Finally we acknowledge conversations with Professor John Blatt and Professor V. F. Weisskopf who pointed out to us the importance in this problem of the condition that the source be bounded.

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Average Charge on the Daughter Atoms Produced in the Decay of A^{37} and Xe^{131m}†

M. L. PERLMAN AND J. A. MISKEL
Chemistry Department, Brookhaven National Laboratory, Upton, New York
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The average charges on the Cl^{37} and Xe^{131} atoms produced in the transitions $A^{37} \rightarrow Cl^{37}$ and $Xe^{131m} \rightarrow Xe^{131}$ are found to be $+3.41 \pm 0.14$ and $+8.5 \pm 0.3$ electron charges, respectively. Estimates of the values to be expected from fluorescent and Auger processes and from Z change are made.

INTRODUCTION

ATTEMPTS have been made to explain the chemical effects which are known to accompany radioactive decay on the basis that the atoms whose nuclei undergo radioactive transition are left in ionized or excited states. In the case of the isomeric transition in Br^{80}, Cooper[1] has calculated that a charge of $+4.7e$ is built up within 10^{-9} second for one K-converted gamma ray. The estimate of De Vault and Libby[2] for Br^{80} is $+7e$. Serber and Snyder[3] have calculated, in the cases of beta decay at $Z=90$ and $Z=10$, average excitations of 125 ev and 54 ev, respectively. Other workers have made calculations of the probability for the ionization of electrons in the various levels as a consequence of beta decay, electron capture, and alpha decay.[4] This probability in the K shell is of the order $1/Z^2$ for electron-capture or beta emission; and the probability changes in shells of higher principal quantum number. Careful measurement has been made of L-electron ejection in the α decay of Po^{210}; the result is about fourteen times larger than theory predicts.[5] The K-electron ejection accompanying the Po^{210} α decay has been measured by Barber and Helm;[6] the experimental results are of the order of magnitude predicted by Migdal.[4] Measurement of K ejection in the β decay of S^{35} gives a result in good agreement with theory.[7] Recently, a preliminary report on the average total charge of the Cl^{37} product of A^{37} decay was published,[8] and some work has been done with compounds of C^{14}, Br^{80}, H^3 and with a Kr–Xe fission product mixture.[9,10] A detailed description of the measurements and results for A^{37} and Xe^{131m} is given in this paper; the experimental method used is free from many of the ambiguities which raise questions about the older charge determinations.

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[1] E. P. Cooper, Phys. Rev. **61**, 1 (1942).
[2] D. De Vault and W. Libby, J. Am. Chem. Soc. **63**, 3216 (1941).
[3] R. Serber and H. S. Snyder, Phys. Rev. **87**, 152 (1952).
[4] A. Migdal, J. Phys. (U.S.S.R.) **IV**, 449 (1941); E. L. Feinberg, J. Phys. (U.S.S.R.) **IV**, 423 (1941); A. Winther, Kgl. Danske Videnskab. Selskab, Mat.-fys. Medd. **27**, No. 2 (1952); J. S. Levinger, Phys. Rev. **90**, 11 (1953); H. M. Schwarz, J. Chem. Phys. **21**, 45 (1953); H. Primakoff and F. T. Porter, Phys. Rev. **89**, 930 (1953).
[5] W. Rubinson and W. Bernstein, Phys. Rev. **86**, 545 (1952).
[6] W. C. Barber and R. H. Helm, Phys. Rev. **86**, 275 (1952).
[7] J. J. Howland, Jr., and W. Rubinson (private communication).
[8] J. A. Miskel and M. L. Perlman, Phys. Rev. **87**, 543 (1952).
[9] S. Wexler and T. H. Davies, Phys. Rev. **88**, 1203 (1952).
[10] Most of the results of older measurements of daughter-atom charge suffered from ambiguities connected with emission of ions from solid sources or with the presence of large background effects. See Mund, Capron, and Jodogne, Bull. soc. chim. Belg. **40**, 35 (1931); G. Hevesy and F. Paneth, *A Manual of Radioactivity* (Oxford University Press, London, 1938); B. Wright, Phys. Rev. **71**, 839 (1947); C. Sherwin, Phys. Rev. **73**, 216 (1948).

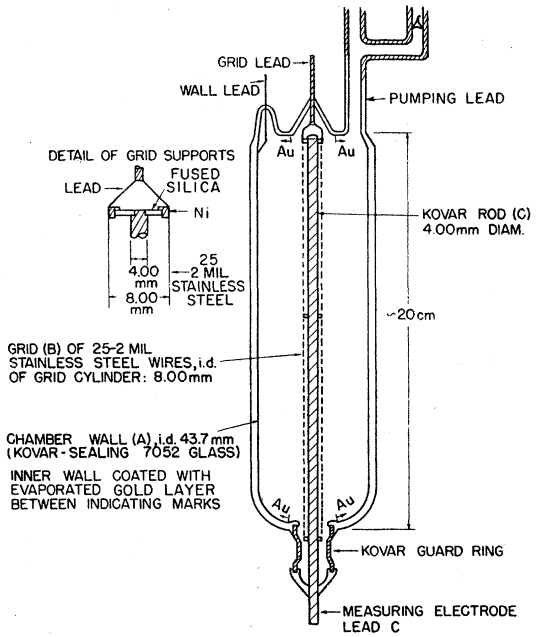

FIG. 1. Chamber for measurement of positive-ion current.

EXPERIMENTAL METHOD AND APPARATUS

The important quantities measured in these experiments were (1) the current resulting from the collection of the primary positive ions produced in a sample of radioactive gas and (2) the disintegration rate of the gas sample. The quotient of the quantities, (1)/(2), is the average charge per disintegration.

Figure 1 is a diagram of the chamber used for the current measurements. The outer electrode A was maintained at any desired positive potential by a stable, quiet power supply;[11] the current produced by the collection of the positive ions at the central electrode C, near ground potential, was measured by a vibrating reed electrometer and Brown recorder. Close to the electrode C and insulated from it by quartz spacers was a grid about 95 percent transparent; the application of a negative potential, battery supplied, to this grid served to return secondary electrons produced at the surface of electrode C to the electrode. Tests showed that electrical leakage from the grid to the collector electrode, at the higest grid voltage used in the experiments, was less than one percent of the smallest measured positive-ion current. The geometry and electric fields are designed to return wall secondaries to the wall and to allow only a small fraction of the energetic primary electrons to intercept the central electrode. Electrode diameters and active and dead volumes of the tube were carefully measured so that the necessary corrections to the measurements could be calculated. Before admission of the radioactive gas to the tube, it was baked and pumped; the A^{37} and Xe^{131m} introduced were essentially carrier-free. It was thus possible, during the course of the current measurements, to maintain in the sealed-off tube a pressure of approximately 10^{-4} mm, corresponding to a mean-free-path of the order of 40 times the tube radius.

By use of the reed electrometer as a null-detector in a Wheatstone bridge, the resistors used with it for the current measurements were calibrated and checked from time to time. The primary standard was a wire-wound resistor, $10^6 \pm 10^3$ ohms. Calibration of the reed-recorder combination was effected by the introduction of accurately known voltages from a Rubicon potentiometer.

When the current measurements on a sample had been completed, the chamber was fused on to a vacuum system at its break-seal. After carrier argon was added, the active gas and carrier were well mixed and transferred, by means of a Toepler pump, to a calibrated part of the vacuum system. By manipulation of this part of the system it was possible to introduce small, known fractions of the active gas into glass proportional counters of a type useful for the determination of absolute disintegration rates.[12] For a given gas sample the results of different assay procedures agreed to approximately plus or minus one percent.

The A^{37} used in these experiments was produced by the n, α reaction in Ca^{40}. Five gram samples of metallic calcium, outgassed at 600°C and sealed in vacuum in silica ampoules, were irradiated for about one month in the Brookhaven reactor. Cadmium foil wrappers served to absorb slow neutrons. The metal samples were put into a silica liner held in a silica tube S, attached to part of a glass vacuum system as shown in Fig. 2. Apiezon grease which had been melted in vacuum was used in the stopcocks. After the metal had been again outgassed at about 700°C, and the current measuring tube T had been given a final bake-out, the cocks A and B were closed; and the calcium was then melted by induction heat. If the vacuum requirement made it necessary, the tube S could be cooled with liquid nitrogen or the barium getter in the side arm could be flashed. Cock B was then opened to allow the active

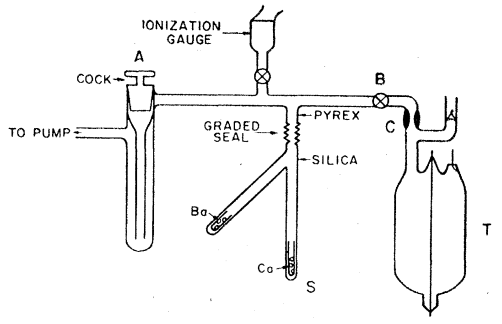

FIG. 2. Vacuum system for filling chamber.

[11] W. A. Higinbotham, Rev. Sci. Instr. **22**, 429 (1951).

[12] W. Bernstein and R. Ballantine, Rev. Sci. Instr. **21**, 158 (1950); S. Katcoff, Phys. Rev. **87**, 886 (1952).

gas to diffuse into T, and then the tube was sealed off at point C and removed.

The Xe^{131m} was the decay product of the chain $Te^{131}\rightarrow I^{131}\rightarrow Xe^{131m}$; irradiation of tellurium metal in the reactor produced the Te^{131}. Except for adjustment of temperatures to take into account the low melting point of tellurium, the method for introduction of the Xe^{131m} into the apparatus was the same as that used for the A^{37}.

MEASUREMENTS AND RESULTS

Measurements of the positive-ion current in the chamber were made over a range of applied wall voltages at each of three grid voltages. Figure 3 shows the results of a set of such measurements for A^{37}. The ordinate in this figure is the voltage developed across a 1.932×10^{10} ohm resistor by the positive-ion current. It is seen that at each grid voltage there is a positive-ion current plateau and that the plateau current is independent of the grid voltage. The current increase at the upper end of the plateaus is caused by the extraction of secondary electrons from the grid-collector space by the positive field from the wall; as expected, the application of increased negative potentials to the grid extends the plateau to higher wall voltages. In each experiment the plateau current decay was followed for at least one half-life as shown in Fig. 4. The half-lives for A^{37} and Xe^{131m} as observed in this way are in good agreement with the accepted values of 34.1 days and 12.0 days, respectively.[13]

The energy of the Xe^{131m} transition is 163 kev. The radiation is 98 percent converted; 63 percent of the conversion occurs in the K shell, 27 percent in the L shell, and 10 percent in the M shell.[14] Activity measurement of the xenon assay samples presented no special difficulties. The unconverted transitions were not detected in the counters and should not have produced current in the chamber.

A^{37} decays by capture of K and L orbital electrons; the L/K ratio is 0.09.[15] In a proportional counter containing A^{37} the pulses fall into two groups, whose relative energies correspond to the binding energies of K and L electrons in chlorine. The A^{37} counters exhibited two plateaus. At the lower voltage plateau only the larger pulses were counted; at the higher plateau both pulse groups registered. The smaller pulses are associated with L-capture events and with those K-capture events which are accompanied by K fluorescence not absorbed in the counter gas. The ratio observed for the count rates on the two plateaus, 1.20 $\pm$0.01, agrees with that calculated from the L/K ratio, the chlorine fluorescence yield[16] (0.11), and the absorption of the counter gas for chlorine K x-rays.

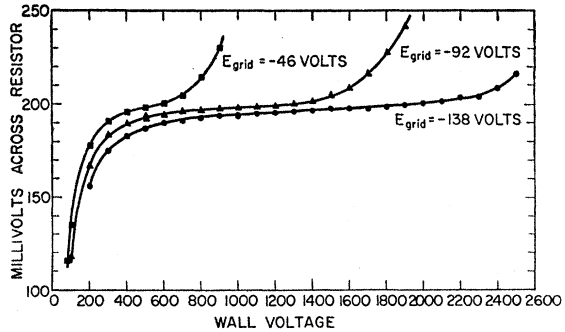

FIG. 3. A^{37} chamber currnt *versus* wall voltage.

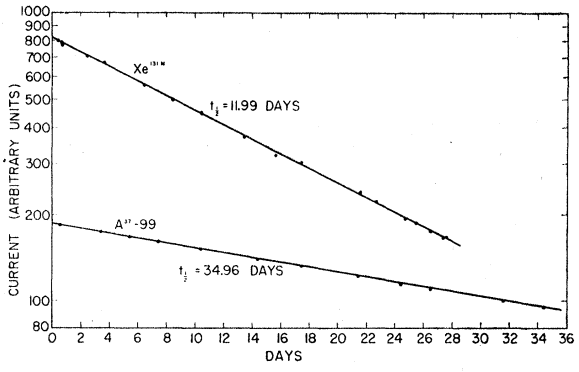

FIG. 4. Decay of A^{37} and of Xe^{131m} chamber currents.

The decay of the A^{37} and Xe^{131m} in the counters was followed for from one to three half-lives. No radioactive impurities were observable.

The chamber currents, sample disintegration rates, and final results are shown in Table I.

The currents tabulated have been corrected to take into account the insensitive volume of the chamber, positive-ion collection efficiency, fast electron-interception by the central electrode, and slow electron-collection by the central electrode. The magnitudes of these corrections applied to the measured currents were approximately 6 percent, 10 percent, 1 percent, and 1 percent, respectively.

The chamber used in the measurement of $A^{37}-1$ was an early model with an aluminum coated wall and glass grid insulators; the larger error given with the $A^{37}-1$ result is related to the existence of grid to central electrode leakage current in this chamber.

DISCUSSION

The average ionic charge expected in the decay of Xe^{131m} may be calculated approximately from the Auger transition probabilities; the recoil energy, less than one ev, is insufficient to produce ionization. For the xenon case the charge was calculated for a K fluorescence yield of 0.87;[16] an L fluorescence yield of 0.11;[17] and

[13] *Nuclear Data* National Bureau of Standards Circular No. 499, (U. S. Government Printing Office, Washington, D. C., 1950).
[14] I. Bergström, Arkiv Fysik **5**, 191 (1952).
[15] Pontecorvo, Kirkwood, and Hanna, Phys. Rev. **75**, 982 (1949).
[16] Broyles, Thomas, and Haynes, Phys. Rev. **89**, 715 (1953).
[17] E. H. S. Burhop, *The Auger Effect* (Cambridge University Press, Cambridge, 1952).

TABLE I. Experimental results

Sample	Chamber current (amperes)	Disintegration rate (events/sec)	Ave. charge per disintegration (electronic charges)	
$A^{37}-1$	1.56×10^{-12}	2.72×10^{6}	$+3.58\pm0.2$[a]	$+3.41\pm0.14$
$A^{37}-2$	4.72×10^{-12}	8.86×10^{6}	$+3.32\pm0.1$	
Xe^{131m}	2.35×10^{-14}	1.72×10^{4}	$+8.5\ \pm0.3$	

[a] This value is obtained by recalculation of the result previously reported (reference 8); an arithmetic error has been eliminated, and the value of the chlorine K fluorescence yield has been changed from 0.08 to 0.11.

division of Auger probability, according to Pincherle,[18] $LL\rightarrow K\infty$ (0.56), $LM\rightarrow K\infty$ (0.32), $LN\rightarrow K\infty$ (0.07), $MM\rightarrow K\infty$ (0.05). It was assumed that an analogous probability division applies to Auger transitions which originate with L level holes. The charge thus calculated, without contribution from Coster-Kronig transitions, is for K converted decay events $+6.72$, for L conversions $+6.62$, and for M conversions 4.00. The average, weighted according to the Xe^{131m} decay scheme, is $+6.41$. It is possible to increase the calculated charge by 3 units by inclusion of some of the energetically allowed Coster-Kronig transitions. Unfortunately, data for the transition probabilities do not seem to be available.

For the A^{37} the division of the Auger probability is $LL\rightarrow K\infty$ (0.65), $LM\rightarrow K\infty$ (0.31), and $MM\rightarrow K\infty$ (0.035). Again without Coster-Kronig contribution, the charge expected in K capture would be 2.42; in L capture it would be 1.0. The weighted average is 2.30. The maximum possible Coster-Kronig contribution to the charge, 0.8, is made if every K hole which is filled by Auger processes involving the L level produces one L_{I} hole and if all L_{I} holes give rise to Coster-Kronig ionizations. L_{I} electrons are expected to be involved in L capture. A rough estimate of the actual Coster-Kronig contribution to the charge for A^{37} would be about one-fifth[19] of 0.8 or 0.16. Thus the experimentally measured charge for A^{37} is at least 0.3 ± 0.14 and more probably 0.9 ± 0.14 electron charges larger than that expected from Auger processes. A change in the division of the Auger probability from that used above to $LL\rightarrow K\infty$ (0.90), $LM\rightarrow K\infty$ (0.09), and $MM\rightarrow K\infty$ (0.01) would add 0.25 unit to the previously calculated charge. In addition there would be an increase in the Coster-Kronig contribution of 0.3 as a maximum or more probably 0.06.

The A^{37} recoil energy, 9.6 ev,[20] is insufficient to cause electron loss.

A small further contribution to the total charge both for A^{37} and Xe^{131m} is to be expected from the change of electrostatic potential in the nuclear transition (electron "shake-off").[4] According to Primakoff[4] the probability of double K hole production in K capture is $3/(4Z^2_{\mathrm{eff}})$, where Z_{eff} is the effective nuclear charge. For A^{37} the contribution from this source to the charge is negligible; ionization in the L and M shells in the K-capture act is also very small because for these electrons the electrostatic potential of a nucleus Z shielded by two K electrons is nearly the same as that of the product nucleus $Z-1$ shielded by one K electron.

It should be pointed out that the foregoing analysis of the "shake-off" process is incomplete. Beta decay, as opposed to K capture, results in a change of electrostatic potential which influences all electrons. The "shake-off" probability here increases with increasing principal quantum number because of the decrease in Z_{eff}. Similarly, the creation of vacancies in the electronic cortege by Auger processes and by internal conversion results in a change of electrostatic potential for all electrons having a principal quantum number equal to or greater than that of the vacancy. This effect should produce an additional charge of approximately 0.05 for A^{37} and approximately 0.2 for Xe^{131m}.

The authors take pleasure in expressing their appreciation to Dr. Max Wolfsberg and to Dr. William Rubinson for valuable discussions.

[18] L. Pincherle, Nuovo cimento **12**, 81 (1935).
[19] Reference 17, p. 77.
[20] Richards, Smith, and Browne, Phys. Rev. **80**, 524 (1950).