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Radioactivity Produced by Proton Bombardment of Bromine and Iodine

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Bromine bombarded with 5–6-Mev protons emits electrons with half-lives 13 ± 2 seconds and 55 ± 2 seconds. A β -spectrograph shows these to be conversion electrons from at least two gamma-rays of energies 127 and 187 kev. Physical and chemical tests indicate the activity is from a krypton isotope. The long period Kr^{79, 81} reported by Snell is found to have a half-life of 34.5 ± 1 hours and a positron upper limit of about 0.4 Mev. Iodine targets similarly emit electrons with a half-life of 75 ± 1 seconds, shown to be conversion electrons from at least two gamma-rays of energies 125 and 175 kev, coming from a xenon isotope. Radiation consisting of electrons, x-rays, and probably gamma-rays, decaying with a half-life of 34 ± 2 days is also found. Secondary electrons produced in the target material or in Pb absorbers by x-rays from the source account for about $\frac{2}{3}$ of the total activity measured by the electroscope.

BROMINE

L EAD bromide, when bombarded with protons of 5-6 Mev energy was found to emit electrons in two radioactive periods of half-lives 13 ± 2 seconds and 55 ± 2 seconds (Fig. 1). The same activities were found in potassium bromide. Since no short periods were found in potassium or lead, these activities must be due to some process taking place in the bromine.

To test whether the element responsible was Br (z=35) or Kr (z=36), the potassium bromide target was dissolved and boiled to dryness, which procedure caused the activity to disappear. A proof that bromine is not volatile under similar conditions was given by bombarding potassium bromide with slow neutrons, forming Br^{80, 82}, and then boiling the target to dryness. Since the activity remained in this case, we believe the 55-second period to be due to a krypton isotope.

Because of its shortness, no definite information could be obtained on the 13-second period, although the indication was that it also is volatile.

A further check for the source of the activity was made with the apparatus shown in Fig. 2. Lead bromide was coated on the nickel foil, and bombarded in vacuum. Solid carbon dioxide and alcohol slush was then put into the small trap, and liquid air in the larger one, while krypton gas flowed into the apparatus. The foil was then heated to vaporize the lead bromide and release any occluded gas. Free bromine should be frozen out by the carbon dioxide, and some of the krypton by the liquid air. The activity on the liquid-air trap was then measured and found to be of about 55-seconds half-life. As a check experiment the carbon dioxide trap was left empty and the CO₂-alcohol mixture put in the liquid-air trap. A bombardment and heating of a new lead bromide target then caused less activity

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FIG. 1. Short periods from bromine bombarded by protons.

to appear on the liquid-air trap than in the previous experiment, with little or no 55-second period measurable.

These experiments make it seem probable that the electrons are from krypton. The stable isotopes of bromine and krypton are shown in Fig. 3. Krypton could be produced in the following ways:

(1)
$$\operatorname{Br}^{79, 81}(p, n)\operatorname{Kr}^{79, 81}$$

(2) Br^{79, 81}
$$(p, \gamma)$$
Kr^{80, 82}.

Because the occurrence of negative electrons suggested an internally converted gamma-ray, a β -ray spectrograph was fitted with a vacuum lock for introducing targets repeatedly until sufficient effect was built up on the photographic film. This lock is described in a note soon to appear in *Review of Scientific Instruments*. One thousand and one bombardments were made to obtain the spectrum. Bombardment time was about one minute, while fifteen seconds elapsed between removing the target from the hydrogen-filled bombarding chamber of the cyclotron and placing it in the spectrograph. Since the intensities of the two periods were about the same immediately after bombardment, the longer should emit approximately four times as many electrons as the shorter. About one-half of the short period electrons were lost while the target was being placed in the spectrograph, while only one-half of the long period electrons were used during the one-minute exposure in the spectrograph. Therefore the lines due to the 55-second period should be about four times as strong as



FIG. 2. Apparatus used to freeze out the radioactive krypton.

those due to the 13-second period. For this reason the stronger two lines may be tentatively assigned to the long period, and the weaker lines to the 13-second period. In Fig. 3, which reproduces the spectrogram, three lines are visible. The very weak fourth line can be seen but poorly on the original. The spectrograph was calibrated with the active deposit of thorium, and on various trials the magnetic field (furnished by an electromagnet and storage batteries) could be reproduced to within $\frac{1}{2}$ percent.

The energies of the electron lines are 0.113, 0.125, 0.173, and 0.185 Mev, respectively. The separation of the K and L_{111} x-ray absorption edges in bromine is 0.0116 Mev, and that in krypton is 0.0124 Mev, while the separation of the two lower energy electron lines is observed to be 0.012, and of the higher two, 0.012 Mev. Because the uncertainty in measuring the lines is about 1 percent, this cannot be considered definite evidence that the lines are from krypton, but if so, the energies of the gamma-rays are:

$$\begin{split} \gamma \ (\text{strong}) &= 0.125 + 0.002 \ (= \text{Kr} \ L_{111} \text{ absorption} \\ &= 0.127 \\ &= 0.113 + 0.014 \ (= \text{Kr} \ K \text{ absorption} \\ &= 0.27 \ \text{Mev.} \\ &= 0.127 \ \text{Mev.} \\ \gamma \ (\text{weak}) &= 0.185 + 0.002 = 0.187 \\ &= 0.173 + 0.014 = 0.187 \ \text{Mev.} \end{split}$$



FIG. 3. Upper photograph: conversion electrons from krypton. Lower photograph: conversion electrons from xenon.

The uncertainty in these values is about 1.5 percent. If the gamma-rays are from bromine, their energies will be lowered by 1 percent, since the K and L electron binding is smaller than in krypton.

Another check on the element was made by studying the x-rays accompanying the 55-second period. No absorbers are available to distinguish Br characteristic x-rays from those of Kr by use of the K-absorption edges, so it was necessary to use the L edges of Au and Pt. A thin layer of lead bromide was melted onto 0.2-mil platinum, and bombarded for two minutes. The 13-second period was allowed to decay for 45 seconds, after which 3 to 4 electroscope readings were

TABLE I. Stable isotopes.Bromine and krypton

	78	79	80	81	82	83	84	85	86		
35Br		50.6		49.4							
36Kr	0.35		2.01		11.53	11.53	57.10		17.47		

Iodine and xenon

	124	125	126	127	128	129	130	131	132	133	134	135	136
53I				100			-						
₅₄ Xe	0.094		0.088		1.90	26.23	4.07	21.17	26.96		10.54	-	8.95



FIG. 5. Short period from iodine bombarded by protons.

made with the absorber in place. The absorber was then removed, and 6 to 8 readings taken. The data suggest that thex-raysare from krypton.

 $\rm Kr^{79,\,81}$ postulated in reaction (1) above is not stable. It has been produced by Snell¹ by deuteron bombardment with a half-life of the order of magnitude of 18 hours. We have bombarded lead bromide and also sodium bromide with protons for about 30 hours and obtained a half-life of 34.5 ± 1 hours which magnetic analysis shows to consist of positrons. See Fig. 4. Examination of these positrons in a cloud chamber reveals an upper energy limit of 0.4 Mev. Scattering in the



FIG. 4. Positron decay of Kr^{79, 81}.

FIG. 6. Absorption of 75-second radiation from I¹²⁷+H¹, showing the x-rays are characteristic of xenon. Total activity was about 1 division/second. Absorption coefficients interpolated from data given in Compton and Allison, X-Rays in Theory and Experiment, are as follows: For iodine characteristic K radiation: in Sn, $\mu/\rho=9$; Ag, 62; Sb, 15. For xenon K radiation: in Sn, 45; Ag, 40; Sb, 10.



gas is rather bad at this energy, however, so the uncertainty in this value is about 0.1 Mev.

¹ A. H. Snell, Phys. Rev. 52, 1007 (1937).

Iodine

The procedure with iodine closely parallels that with bromine. The stable isotopes, and those of xenon, are shown in Table I. When lead iodide, potassium iodide, and lead on which iodine had been melted and recrystallized were bombarded with protons, an activity of 75 ± 1 seconds was observed (Fig. 5). This activity was volatile, while I¹²⁸ formed by slow neutron capture was not volatile under similar conditions.

Two β -ray spectrograms were made of the electrons, one using 801 lead iodide targets, and one using 1301 potassium iodide targets. Figure 3 shows that obtained from lead iodide which differs in no important respect from the other. Three lines are visible, at 0.0914, 0.140, and 0.170 Mev, respectively. The $K-L_{111}$ separation in xenon is 0.029 Mev, while that in iodine is 0.0278 Mev. Here, as in the case of bromine, the error is too great to assign definitely the observed separation (0.170-0.140=0.030) of the upper two lines to either element. The lowest energy



FIG. 8. Absorption of 34-day period from $I^{127}+H^1$. Curves A and B show absorption in aluminum. Curve C was made with lead absorbers but with 31 mg/cm² of aluminum between the absorber and the electroscope to remove the soft secondary radiation produced in the lead. Total activity was about 2 divisions/minute.



FIG. 9. Absorption of 34-day period from $I^{127}+H^1$. The filled-in circles show the absorption by lead alone; the open circles, by aluminum alone. The points marked x were obtained by putting various thicknesses of aluminum between 318 mg/cm² of lead and the electroscope. The points marked + show the activity observed through lead absorbers, but with 31 mg/cm² of aluminum absorbing the secondary electrons produced in the lead.

line appears to be single, although one several times weaker could be lost in the background. If this is assumed to be a K line from xenon, the gamma-ray energy is:

0.0915 + 0.034 (Xe K-absorption edge) = 0.125 Mev.

The higher energy gamma-ray is, similarly,

0.140 + 0.034 = 0.174

or 0.170 + 0.005 (Xe L_{111} -absorption edge) = 0.175 Mev.

The uncertainty in these values is about 1.5 percent. A chemical investigation showed that the amount of bromine in the lead iodide, or iodine in the lead bromide, if any at all, was much too small to account for the coincidence in energy of the two gamma-rays (Br, 127 kev and I, 125 kev).

An attempt was made to freeze out the active material with liquid air, but the results were less positive than in the case of bromine, which is perhaps not surprising, as the vapor pressure of xenon at liquid-air temperature is about 13 centimeters, and only a small amount was used as a carrier. However, the volatility of this activity makes it seem that a xenon isotope is responsible. Furthermore, the x-rays accompanying the internal conversion were shown to be characteristic of xenon. See Fig. 6. A more complete study of the absorption in silver showed the ionization due to the x-rays was about equal to that of the unconverted gamma-rays.

The possible ways of producing xenon are the following:

(3)
$$I^{127}(p, n)Xe^{127}$$

(4)
$$I^{127}(p, \gamma)Xe^{128}$$
.

Xe¹²⁸ is stable, and the gamma-rays may arise from an excited state in it. Xe¹²⁷ is not found in nature, nor has it been reported as a radioactive isotope. We have bombarded lead iodide and also sodium iodide for about 30 hours and obtained an activity of 34 ± 2 days half-life (see Fig. 7), consisting of electrons, x-rays and probably some gamma-rays. The range of the electrons is about 350 mg/cm² of aluminum. By means of a counter kindly supplied by Mr. J. G. Fox, they were found to be negative. A softer (negative) group also appears in the complete absorption curve (Fig. 8), with range about 35 mg/cm² of aluminum. These, however, are undoubtedly secondaries produced by the harder radiation in the material of the source and the absorber, since the same group is found by absorbing in aluminum the radiation that comes through various thicknesses of lead. The greater number of these secondaries produced in lead than in aluminum makes the total radiation received through a given mass of lead greater than that through the same mass of aluminum, until the total absorber thickness is about 0.9 gram/cm². This effect is shown in Fig. 9.

A study of the x-radiation with critical absorbers in an attempt to ascertain whether the x-ray is from iodine or xenon gave an inconclusive result. From the absorption coefficient in Pb, its energy is found to be about 40–45 kev.

Although the number of nuclei with the 75second half-life made in a given bombardment is of the same order of magnitude as the number decaying with the 34-day period, no genetic relationship between the two processes has as yet been established.

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Internal Conversion of Gamma-Radiation in the L Shell

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The nonrelativistic calculations of the internal conversion of electric multipole radiation which have been made for the K shell are extended to the L shell. Numerical values of the conversion coefficient for both the K and L shells are given and curves showing the ratio of the K conversion to the L conversion for different energies and atomic number are presented. This latter ratio is quite sensitive to the multipole order and varies between ~ 0.1 and 10 in the range in which the calculations are valid. A simple relativistic formula for the conversion of magnetic multipole radiation is given. In Section 5 we summarize selection rules and give applications of the formulas to experiments.

1. INTRODUCTION

THE increasing number of cases in the lighter elements in which the internal conversion of gamma-radiation has been observed makes a study of this phenomenon more important. Calculations of the ratio of the number of conversion electrons to the number of gamma-quanta N_e/N_q



FIG. 3. Upper photograph: conversion electrons from krypton. Lower photograph: conversion electrons from xenon.