

Since the cascade from the 209-keV level to the 119-keV level is extremely weak, Fig. 4 represents the yield of the neutron group leading to the 119-keV level. The qualitative appearance of this curve, including pronounced resonances, is very similar to the total neutron yield from the  $\text{Cu}^{65}(p,n)\text{Zn}^{65}$  reaction measured<sup>10</sup> with a  $\text{BF}_3$  counter.

#### D. NEUTRON BRANCHING RATIOS

Using the data in Table II, one can calculate the neutron branching ratios to the first three excited states. Sufficient data to do this were taken at three bombarding energies; 3 MeV, 3.75 MeV, and 4 MeV. The results are given in Table III. The experimental uncertainty in the relative intensities is 25%. From the present work nothing can be said about the relative intensity of the neutrons leading to the ground state. However, the previous work<sup>6</sup> at Rice showed that at 3 MeV the groups involving the 119-keV level and the ground state have approximately the same intensity.

<sup>10</sup> Brugger, Bonner, and Marion, *Phys. Rev.* **100**, 84 (1955).

TABLE III. Branching ratios for the neutron groups.

Level (keV)	Neutron group <sup>a</sup>	Relative intensity <sup>b</sup>		
		3.00 MeV	3.75 MeV	4.00 MeV
0	<i>n</i>	8 <sup>c</sup>	...	...
54	<i>n'</i>	...	1	...
119	<i>n''</i>	8	8	8
209	<i>n'''</i> <sup>d</sup>	3.8 (3.2)	4 (3.5)	3 (2.7)

<sup>a</sup> See Fig. 2.

<sup>b</sup> Arbitrarily normalized to 8 for *n''* at each energy.

<sup>c</sup> From reference 5.

<sup>d</sup> See Table II, footnote d.

#### DISCUSSION

The experiment described illustrates quite nicely the advantages of the rather new technique for studying nuclear reactions. Except for the spins, a rather complete description of the first three excited states of  $\text{Zn}^{65}$  is obtained. The existence of the level at 54 keV which is directly excited by neutrons severely limits the  $\text{Cu}^{65}(p,n)\text{Zn}^{65}$  reaction as a source of neutrons. This limitation was suggested from the previous neutron work.<sup>6</sup>

### Ionization Following Beta Decay in Krypton-85

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Following beta decay in the 9.4-year radioactivity of  $\text{Kr}^{85}$ , the product rubidium atoms are found to be distributed in the following charge spectrum (figures in percent): charge 1,  $79.2 \pm 1.0$ ; charge 2,  $10.9 \pm 0.2$ ; charge 3,  $3.9 \pm 0.1$ ; charge 4,  $3.1 \pm 0.1$ ; charge 5,  $1.51 \pm 0.07$ ; charge 6,  $0.66 \pm 0.04$ ; charge 7,  $0.40 \pm 0.02$ ; charge 8,  $0.19 \pm 0.02$ ; charge 9,  $0.093 \pm 0.01$ ; charge 10,  $0.026 \pm 0.006$ ; charge 11, unobservable. An interpretation is made in terms of the "shaking off" of extranuclear electrons by the sudden change by +1 in the nuclear charge, the intensity of the charge-2 ions being taken as a direct measure of the probability of "shake-off" from the combined 4*s* and 4*p* shells. The combined intensities of charge 3 and higher are correspondingly equated to the summed probabilities of primary ionizations in the  $n=1, 2$ , and 3 shells.

THE atomic-charge spectra that result from radioactive decay appear to have a variety and an interest in their own right, reflecting a sensitivity to the nature of the nuclear change that caused the ionization, as well as representing the statistical outcome of the complex atomic rearrangements that follow electron loss from any of the several electron shells of the atom. We have already presented the peaked charge distribution that results from a single internal-conversion event in the 12-day isomer of  $\text{Xe}^{131}$ ,<sup>1</sup> and the similarly peaked distribution produced by electron capture in  $\text{A}^{37}$ .<sup>2</sup> The peaked distributions are caused by vacancy cascades that follow the creation of an electron hole in one of the inner shells. By way of contrast, we

wish now to describe a charge spectrum that follows pure beta emission, and we shall see that it consists of a monotonic decrease, sharply falling toward the higher charges. The radionuclide is  $\text{Kr}^{85}$ , chosen as one in a series that give information uncomplicated by molecular effects.

Although the qualitative comparison of the different kinds of charge spectra is of interest, there is a further significance in the present study, for it provides a new kind of opportunity—and experimentally apparently a good opportunity—for a check of the theory of electron shake-off following an abrupt change in the nuclear charge. It will be recalled that with the rapid escape of the beta particle in a time short compared with the orbital times of the electrons, the change in the nuclear charge from  $Z$  to  $Z \pm 1$  introduces a sudden perturbation into the electrostatic environment of the electrons, with a resulting possibility of excitation or ionization. The process has been considered on a rather general

<sup>1</sup> F. Pleasonton and A. H. Snell, *Proc. Roy. Soc. (London)*. (to be published); A. H. Snell and F. Pleasonton, *Phys. Rev.* **102**, 1419 (1956).

<sup>2</sup> A. H. Snell and F. Pleasonton, *Phys. Rev.* **100**, 1396 (1955); see also O. Kofoed-Hansen, *Kgl. Danske Videnskab. Selskab, Mat-fys. Medd.* **29**, 15 (1955), and *Phys. Rev.* **96**, 1045 (1954).

theoretical basis by Feinberg,<sup>3</sup> Migdal,<sup>4</sup> Levinger,<sup>5</sup> Schwartz,<sup>6</sup> and Benoist-Gueutal<sup>7</sup> and more specialized contributions to the subject have been made by Winther and Kofoed-Hansen<sup>8</sup> and by Primakoff and Porter.<sup>9</sup> The probability of ionization is found to be of the order  $1/Z_{\text{eff}}^2$  per electron, and the calculations so far have extended only to the  $K$ ,  $L$ , and  $M$  shells. Experimental confirmation has been sought by absolute counting of the x-rays that follow ionization from the inner shells. The results have supported the theory reasonably well over a wide range of atomic number; we may cite the measurements of Renard<sup>10</sup> in  $\text{P}^{32}$ , of Rubinson and Howland<sup>11</sup> in  $\text{S}^{35}$ , of Michalowicz and Bouchez<sup>12</sup> in  $\text{Sr}^{90}$ — $\text{Y}^{90}$ , of Boehm and Wu<sup>13</sup> in  $\text{Pm}^{147}$ , and several studies<sup>10,12,13</sup> of  $\text{RaE}$  as pertinent examples. It is clear, however, that despite their value in checking the theory, measurements of this kind nevertheless miss the main effect because screening reduces  $Z_{\text{eff}}$  so sharply that most of the ionizations will in fact take place from the *outer* electron shells. It therefore is revealing to turn the attention of both experiment and theory to the exterior part of the atom.

The application of charge spectrometry to the product atoms suggests a way of accomplishing this from an experimental point of view. In the case of beta-minus emission, singly-charged positive ions result from the loss of the beta particle alone, while atoms that have in addition lost a single orbital electron will of course appear as charge 2. A consideration of fluorescence yields and line intensities in the various x-ray series shows that if the primary ionization were to take place in any shell other than the outer shell, the probability of at least one subsequent Auger event occurring at some stage in the reorganization of the atom is so large (about 0.99 for Rb) that the atom will almost surely be thrown into a state of ionization higher than 2. The situation suggests therefore that the relative intensity of charge-2 ions in the spectrum will represent almost exactly the probability of shake-off from the outermost shell. Experimentally this leads to a neat situation in which absolute counting is unnecessary, and all that one has to do is to compare the intensities of spectrometer lines. Correspondingly, the combined intensities in charges 3 and higher in the total charge spectrum will give the sum of the probabilities of shake-off for all shells except the outermost, and an additional comparison with theory becomes possible.

<sup>3</sup> E. L. Feinberg, J. Phys. U.S.S.R. 4, 424 (1941).

<sup>4</sup> A. Migdal, J. Phys. U.S.S.R. 4, 449 (1941).

<sup>5</sup> J. S. Levinger, Phys. Rev. 90, 11 (1953) and J. phys. radium 16, 556 (1955).

<sup>6</sup> H. M. Schwartz, J. Chem. Phys. 21, 45 (1953).

<sup>7</sup> P. Benoist-Gueutal, Ann. phys. 8, 593 (1953).

<sup>8</sup> A. Winther and O. Kofoed-Hansen, Kgl. Danske Videnskab. Selskab, Mat-fys Medd. 27, No. 14 (1953).

<sup>9</sup> H. Primakoff and F. T. Porter, Phys. Rev. 89, 930 (1953).

<sup>10</sup> G. A. Renard, J. phys. radium 16, 575 (1955).

<sup>11</sup> W. Rubinson and J. J. Howland, Phys. Rev. 96, 1610 (1954).

<sup>12</sup> A. Michalowicz and R. Bouchez, J. phys. radium 16, 578 (1955).

<sup>13</sup> F. Boehm and C. S. Wu, Phys. Rev. 93, 518 (1954).

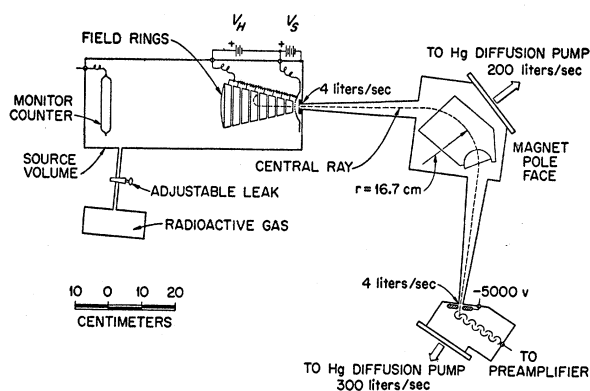


FIG. 1. Diagram of the major components of the charge spectrometer.

The disintegration scheme of the 9.4-year activity in  $\text{Kr}^{85}$  has been studied rather thoroughly.<sup>14</sup> More than 99% of the nuclei decay by beta emission to the ground state of  $\text{Rb}^{85}$ . The maximum energy for the beta particles is 0.69 Mev, and the corresponding maximum recoil energy would be 8.0 ev. There is also a very weak branch in which the beta emission leads to an excited state at 0.51 Mev in  $\text{Rb}^{85}$ . The associated gamma ray has been found by Zeldes, Ketelle, and Brosi,<sup>15</sup> who estimated that 0.65% of the disintegrations follow this path. This may, however, be an overestimate, because at the time of the observation it was not known that the gamma ray is delayed with a half-life of 0.9 microsecond; thus the coincidence rate was probably too low, and the derived intensity figure too high. At any rate, no sign of a conversion line appears in either of the two published beta spectra.<sup>15,16</sup> The 0.51-Mev level is also known in the electron-capture decay of  $\text{Sr}^{85}$ , and the internal-conversion coefficient has been measured by Sunyar *et al.*<sup>17</sup> and by Emmerich and Kurbatov,<sup>18</sup> the results agreeing at the value  $7 \times 10^{-3}$ . It therefore seems clear that internal conversion takes place in fewer than  $10^{-4}$  of the disintegrations, and hence cannot perturb our charge spectrum to any appreciable extent.

## EXPERIMENTAL

The charge spectrometer is sketched in Fig. 1. The ion source was a vacuum vessel containing the radioactive gas at a low pressure (less than  $1.2 \times 10^{-5}$  mm Hg total) such that mean free paths were long compared with the size of the apparatus. In the vessel there was a series of ring electrodes, arranged as an interrupted cone, with a potential  $V_H$  so divided among them that positive ions formed within the cone would be focused toward a location that was also the object position for a

<sup>14</sup> S. Thulin, Arkiv Fysik 9, 162 (1955).

<sup>15</sup> Zeldes, Ketelle, and Brosi, Phys. Rev. 79, 901 (1950).

<sup>16</sup> I. Bergstrom, Arkiv Fysik 5, 191 (1952).

<sup>17</sup> Sunyar, Mihelich, Scharff-Goldhaber, Goldhaber, Wall, and Deutsch, Phys. Rev. 86, 1023 (1952).

<sup>18</sup> W. S. Emmerich and J. D. Kurbatov, Phys. Rev. 85, 149 (1952).

two-directional focusing magnetic analyzer. Here the ions were subjected to a further acceleration  $V_S$  which was made 24 times as large as  $V_H$ . Thus the collecting electrodes were biased relative to ground by a strong positive voltage (400 to 4800 volts in this experiment), and the exact point of origin of an ion within the collecting volume was of little moment in determining its energy at the entrance to the analyzer. When the instrument was properly stabilized and calibrated, a single setting was sufficient to transmit nearly the entire intensity of a given line in the charge spectrum; one was not required to sweep over each ion peak in detail, and the charge spectrum could be surveyed line by line in a succession of widely-separated adjustments.

After deflection through the magnet, the ions were further accelerated through 5 kv for counting by means of an electron multiplier. Differential pumping as indicated in the figure kept the radioactive gas away from the neighborhood of the detector.

The efficiency of collection was kept uniform from charge to charge by applying collecting and accelerating voltages that were inversely proportional to the charge of the ions to be counted. So long as this is the case, both the electric and the magnetic forces upon the ions should be gratifyingly uniform, but on the other hand the requirement may lead to an unnecessarily inefficient collection of the ions of high charge. A handicap of this kind was circumvented by surveying the charge spectrum in overlapping groups of lines. Starting with the magnet at full power, charge 1 was measured, after which charge 2 was counted at half the applied voltage and an appropriately lower magnetic field. Charge 2 was then repeated at maximum magnetic field strength and increased voltage, and considerably more intensity was registered because the collecting field was stronger *vis a vis* the recoil energy of the ions. This second group of lines was usually completed by taking charge 3 at  $\frac{2}{3}$ , and charge 4 at  $\frac{1}{2}$  of the voltage used for charge 2. Then the fields were increased again, and a third group was started by repeating charge 4. After all the groups were covered, they were related to one another to yield the complete spectrum. This procedure gives a system that provides increasing efficiency of collection as one goes toward the higher charges, and as will be seen it enabled us to cover a wide intensity range. The source strength was monitored continuously by a counter shown in the figure.

The ions of low charge were slightly less efficient than those of high charge in producing counts in the multiplier. The associated corrections were evaluated by taking integral bias curves using the Kr ions themselves; they were all upward corrections and amounted to 10% for charge 1, 7% for charge 2, 4.4% for charge 3, and 2.0% for charge 4; they were negligible for ions of higher charge.

The  $\text{Kr}^{85}$  was kindly supplied to us by Dr. G. W. Parker, of the Chemistry Division of this Laboratory. It was fission-product gas, aged three years or more,

separated from xenon and radioactively pure so far as we could see, although it was mixed with stable fission-product krypton isotopes. The rather long half-life meant moderately low counting rates, typical values being 840 counts per minute for charge 1, and 5 counts per minute for charge 10 (in a different group). Background rates were about 20 counts per minute with the radioactive gas present; they were measured separately for each peak by leaving the magnet on the peak setting, but reversing the potential  $V_H$  across the collecting rings.

### EXPERIMENTAL RESULTS

Although it was unnecessary to trace out the peak associated with each charge state in detail, still it was instructive to inspect a line profile occasionally, as an indication of the performance of the spectrometer and as a test of contamination by ions of neighboring mass. Figure 2 is such a profile of the charge-7 ions, obtained actually with a proportionately larger collecting voltage  $V_H$  than was usually used. The full width at half maximum is 1.7% in momentum, as compared with 1.4% for the transmission curve of the spectrometer. The potential drop across the collecting volume apparently did not appear sensitively in the line width.

Because of the rather low specific activity of this gas, it was necessary to use rather higher pressures in the collecting volume than would have been ideal. We investigated the effect of pressure by obtaining curves such as are presented in Fig. 3. Here the effect of collisions in degrading the charge spectrum is evident; the ions of low charge increase in number as the pressure is raised, and those of high charge decrease. An investi-

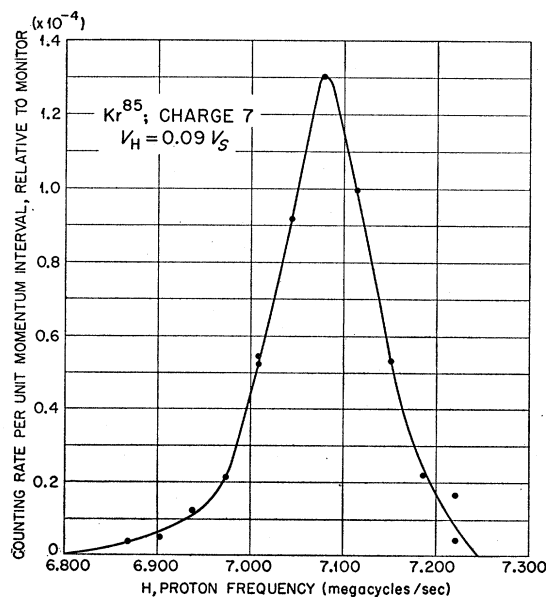


FIG. 2. The profile of the charge-7 line, reproduced as an example of the functioning of the spectrometer and to illustrate freedom from contamination by ions of neighboring charge or mass. One megacycle per second corresponds to 3920 gauss-cm.

gation of this kind enabled us to estimate pressure corrections when they had to be applied to the measured intensities, and the intensities thus corrected are presented in Table I as what we think are our most reliable figures for the charge spectrum. The figures are presented also in the semilogarithmic histogram of Fig. 4. For comparison we also give in Table I the relative intensities before application of the pressure correction.

The errors quoted in the table are based partly upon the statistical errors of counting, and partly upon our feeling of the uncertainty in the pressure corrections and the multiplier sensitivity corrections that were applied. The charge distribution repeated quite accurately under varying conditions, and in measurements that were widely separated in time. The mean charge as derived from the differential distribution is  $1.42 \pm 0.03$ .

### DISCUSSION

In the remarks that follow, we shall take the position that it is energetically impossible in  $\text{Rb}^{++}$  for a 4s vacancy to be filled by a radiationless transition. Numerically, this amounts to saying that the 4s-4p energy difference is less than the ionization energy of  $\text{Rb}^{++}$ , viz. 39.7 ev,<sup>19</sup> and the table of Hill, Church, and Mihelich<sup>20</sup> suggests the truth of the assertion in this region of the elements. It follows that a primary shake-

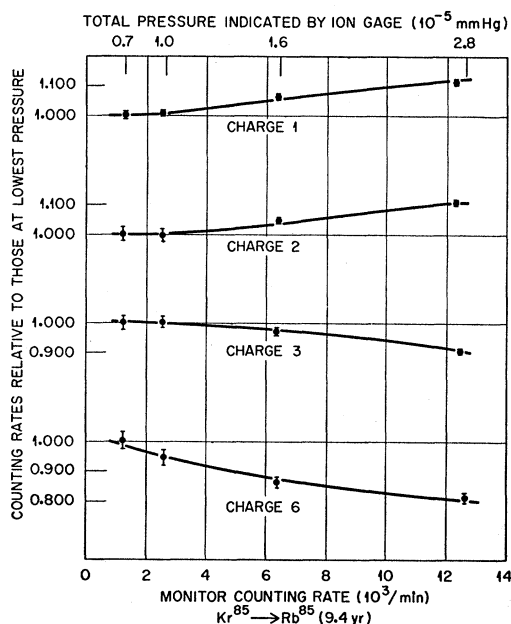


FIG. 3. Effect of pressure upon the measured intensities of some typical ions in the charge spectrum of  $\text{Kr}^{85}-\text{Rb}^{85}$ . Total pressures in the source volume are shown in the upper scale; the radioactive gas was not pure  $\text{Kr}^{85}$  inasmuch as it contained stable fission-product Kr isotopes. 1000 counts per minute on the monitor corresponded to the addition of Kr gas to a partial pressure of  $1.1 \times 10^{-6}$  mm Hg.

<sup>19</sup> G. Joos and A. Saur, Landolt-Bornstein Tables (Springer-Verlag, Berlin, 1950), sixth edition, Vol. 1, Part 1, p. 174.

<sup>20</sup> Hill, Church, and Mihelich, Rev. Sci. Instr. 23, 523 (1952).

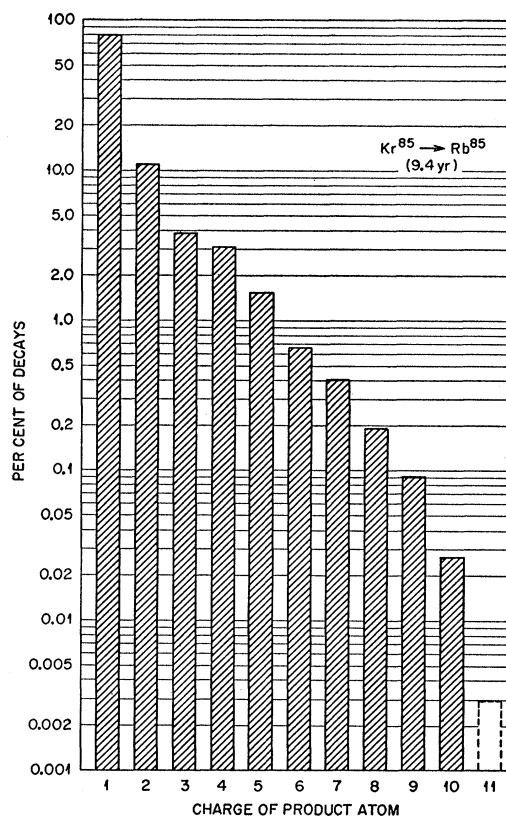


FIG. 4. The charge spectrum of Rb ions formed as the result of beta decay in the 9.4-year activity of  $\text{Kr}^{85}$ .

off of either a 4s or a 4p electron will yield charge 2 ions, but will yield no ions of higher charge.

On this basis we can now proceed to support the assertion that was made early in this paper, to the effect that almost any primary ionization that takes place in the  $n=1, 2$  or 3 shells will throw the atom into a charge state higher than 2. This can be done by taking an example, so let us suppose that a primary vacancy occurs through the shake-off of a  $2p_{3/2}$  electron, and inquire as to the probability  $R(2p_{3/2})$  that the vacancy be filled by a sequence of purely radiative transitions. This probability is first of all proportional to the fluorescence yield  $w(2p_{3/2})$  of the subshell in question; there is then a choice of x-ray transitions, the relative

TABLE I. Charge spectrum of ions following 9.4-year  $\text{Kr}^{85}-\text{Rb}^{85}$  decay.

Charge	Intensity: percent of total		Intensity: percent of total	
	Without pressure correction	With pressure correction	Without pressure correction	With pressure correction
1	79.61	$79.2 \pm 1.0$	7	$0.35$
2	10.93	$10.9 \pm 0.2$	8	$0.17$
3	3.85	$3.91 \pm 0.12$	9	$0.078$
4	2.99	$3.12 \pm 0.13$	10	$0.021$
5	1.41	$1.51 \pm 0.07$	11	$< 0.002$
6	0.60	$0.66 \pm 0.04$		$< 0.003$

probabilities  $I(x-2p_{\frac{1}{2}})$  of which correspond to the relative intensities of the x-ray lines associated with electron transitions toward the  $2p_{\frac{1}{2}}$  vacancy. If the vacancy is carried only as far as the next shell, the fluorescence yield associated with its new location is involved. Taking into account the various allowed transitions, we can accordingly write

$$R(2p_{\frac{1}{2}}) = w(2p_{\frac{1}{2}}) \{ I(3s-2p_{\frac{1}{2}})w(3s) + I(4s-2p_{\frac{1}{2}}) \\ + I(3d_{\frac{3}{2}}-2p_{\frac{1}{2}})w(3d_{\frac{3}{2}}) \},$$

where the three probabilities  $I$  are normalized so that their total is unity. Corresponding expressions can be set up for all of the  $n=1, 2$ , and  $3$  subshells in  $\text{Rb}^+$ , but to evaluate the various  $R$  probabilities accurately one would have to know the fluorescence yields and x-ray intensities for all of the subshells. Much of this information is unavailable, and we can at best make estimates. For the fluorescence yields, we have from Broyles, Thomas, and Haynes<sup>21</sup> and from Burhop<sup>22</sup>  $w(1s)=0.65$  as a firm figure, and although Burhop<sup>23</sup> suggests for the  $n=2$  shell values of the order  $10^{-3}$ , we shall endeavor to be conservative by taking  $w(2s)=10^{-2}$  and  $w(2p_{\frac{1}{2}})=w(2p_{\frac{3}{2}})=2 \times 10^{-2}$  as more in line with the discussion of Robinson and Fink.<sup>24</sup> [The lower value for  $w(2s)$  is a crude recognition of the energetic possibility of Coster-Kronig transitions, which would depress  $w(2s)$  relative to  $w(2p_{\frac{1}{2}})$  and  $w(2p_{\frac{3}{2}})$ .] For the  $n=3$  shell, we follow Burhop,<sup>23</sup> again bearing in mind the possibility of Coster-Kronig transitions, and adopt  $w(3s)=w(3p_{\frac{1}{2}})=w(3p_{\frac{3}{2}})=5 \times 10^{-4}$ , and  $w(3d_{\frac{3}{2}})=w(3d_{\frac{5}{2}})=10^{-3}$ . For the intensities of the x-ray lines we have relied mainly on the summary discussion given by Compton and Allison.<sup>25</sup> In the  $K$  series we have  $I(2p_{\frac{1}{2}}-1s)=0.26$ ,  $I(2p_{\frac{3}{2}}-1s)=0.52$ ,  $I(3p_{\frac{1}{2}}-1s)=0.066$ ,  $I(3p_{\frac{3}{2}}-1s)=0.13$ , and  $I(4p_{\frac{1}{2}, \frac{3}{2}}-1s)=0.018$ . Direct information on the relative intensities of  $L$ -series lines in  $_{37}\text{Rb}$  is scarce, and one is forced to adopt the results for  $_{42}\text{Mo}$ , with a few excursions to even heavier elements. For transitions to the  $2s$  level we have taken  $I(3p_{\frac{1}{2}}-2s)=0.34$ ,  $I(3p_{\frac{3}{2}}-2s)=0.50$ ,  $I(4p_{\frac{1}{2}}-2s)=0.064$ , and  $I(4p_{\frac{3}{2}}-2s)=0.096$ ; to the  $2p_{\frac{1}{2}}$  level  $I(3d_{\frac{3}{2}}-2p_{\frac{1}{2}})=0.97$ ,  $I(3s-2p_{\frac{1}{2}})=0.02$ , and  $I(4s-2p_{\frac{1}{2}})=0.01$ ; and to the  $2p_{\frac{3}{2}}$  level  $I(3d_{\frac{5}{2}}-2p_{\frac{3}{2}})=0.85$ ,  $I(3d_{\frac{3}{2}}-2p_{\frac{3}{2}})=0.10$ ,  $I(3s-2p_{\frac{3}{2}})=0.03$ , and  $I(4s-2p_{\frac{3}{2}})=0.009$ . Fortunately we do not need relative intensities for the  $M$  series, because of the equivalence against radiationless transitions of the  $4s$  and  $4p$  shells.

Using the above information, one can calculate the values of the various  $R$  probabilities. The resulting

estimates are as follows:

$$R(3d_{\frac{3}{2}}) = R(3d_{\frac{5}{2}}) = 10^{-3}, \\ R(3s) = R(3p_{\frac{1}{2}}) = R(3p_{\frac{3}{2}}) = 5 \times 10^{-4}, \\ R(2s) = 2 \times 10^{-3}, \quad R(2p_{\frac{1}{2}}) = R(2p_{\frac{3}{2}}) = 2 \times 10^{-4}, \\ R(1s) = 1.2 \times 10^{-2}.$$

The figures show rather strikingly how seldom an atom readjusts itself without radiationless transitions. It is also apparent that for the most part our guesses as to fluorescence yields and line intensities did not have to be very accurate to show how few inner-shell vacancies lead to charge-2 ions. The reason for the association of the highest probability in the list with the  $1s$  shell derives from the rather high  $1s$ -shell fluorescence yield, coupled with the possibility of a  $(4p-1s)$  x-ray transition which jumps over intervening shells with their paralyzingly small fluorescence yields.

It appears therefore that even if the probability of shake-off were uniform from shell to shell, only one or two percent of the inner-shell shake-offs would lead to charge-2 ions. Actually the  $1/Z_{\text{eff}}^2$  factor and the electron populations assure that  $1s$ -shell primary ionizations will be rarer than  $4s$ - or  $4p$ -shell ionizations by a factor of about 20; it therefore seems clear that primary ionizations in other than the  $4s$  and  $4p$  shells will lead to charge-2 ions so seldom that they will be seen to a negligible extent in this experiment. The primary ionizations in the inner shells will, on the other hand, give rise to vacancy cascades, and it is they that are responsible for the ions of charge 3 and higher.

These considerations have of course failed to take into account the possibility of the simultaneous excitation of two or more electrons. "Autoionization" is known<sup>26</sup> in  $\text{Kr}$ , which is isoelectronic with  $\text{Rb}^+$ , and doubtless some charge-2 ions can be produced when two of the  $\text{Rb}^+$  electrons are simultaneously excited to appropriate bound states, such that one, in returning to the ground state, sends the other into the continuum. It is hard to be certain that this process is unlikely in comparison with the direct loss of a single electron, but one might judge it to be so on the basis of the larger number of final states that are available in the continuum as compared with the number of autoionizing bound states, with the knowledge that plenty of energy is available from the beta decay. Furthermore, if the 20.8% of ionizations found in this experiment is even roughly indicative of the frequency of single-electron excitation, and if the excitation of a first and a second electron are independent events, one might again expect that two-electron excitation might be relatively infrequent. Clearly experiments of a different kind would be required to settle these questions in detail.

In summary, the measurements and their interpretation lead to the conclusion that in the  $\text{Kr}^{85}-\text{Rb}^{85}$  decay, there is a probability of 10.9% that a  $4s$  or a

<sup>21</sup> Broyles, Thomas, and Haynes, *Phys. Rev.* **89**, 715 (1953).

<sup>22</sup> E. H. S. Burhop, *The Auger Effect and Other Radiationless Transitions* (Cambridge University Press, Cambridge, 1952), p. 48.

<sup>23</sup> E. H. S. Burhop, *J. phys. radium* **16**, 625 (1955).

<sup>24</sup> B. L. Robinson and R. W. Fink, *Revs. Modern Phys.* **27**, 424 (1955).

<sup>25</sup> A. H. Compton and S. K. Allison, *X-rays in Theory and Experiment* (D. Van Nostrand and Company, Inc., New York, 1935), p. 637.

<sup>26</sup> H. E. White, *Phys. Rev.* **38**, 2016 (1931).

4*p* electron will be excited to the continuum as a result of the nuclear charge alteration, and that the summed probabilities for like processes in the other shells amount to 9.9%. An extension to the outer electron shells of the calculations based upon the sudden-perturbation

theory is being undertaken by A. E. S. Green, and preliminary results appear to indicate a reasonably satisfactory agreement with the numbers just cited. Dr. Green will doubtless communicate his complete results in the near future.

## Precision Determination of the Low-Lying Energy Levels of W<sup>182</sup>, W<sup>183</sup>, W<sup>184</sup>, and W<sup>186</sup>†

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The low-lying energy levels in the wolfram isotopes have been studied by using the 3.7-Mev proton beam of the A-48 high-current linear accelerator at the University of California Radiation Laboratory, Livermore, to excite the nuclei. The large beam currents available make it possible to use a bent-quartz-crystal spectrograph to observe the resultant  $\gamma$  rays. With such an instrument, a resolution of better than 0.1% is possible. The following  $\gamma$  rays have been observed: W<sup>182</sup>, 100.07±0.05 kev; W<sup>183</sup>, 46.508±0.030 kev and 52.612±0.030 kev; W<sup>184</sup>, 111.13±0.06 kev; W<sup>186</sup>, 122.48±0.08 kev. (Energies relative to W *K*-series x-rays.) The isotopic assignments are known from previous work on these lines. Known wavelengths of the *K*-series x-rays of wolfram and certain nuclear lines were used to calibrate the plate.

### I. A-48 ACCELERATOR

THE A-48 accelerator<sup>1</sup> at the University of California Radiation Laboratory, Livermore is a resonant-cavity linear accelerator designed to accelerate large quantities of protons to 3.7 Mev and deuterons to 7.5 Mev. The maximum current of both species of particles which has been obtained so far is roughly 30 milliamperes. The energy spread of the beam is of the order of several hundred kilovolts because of the large phase acceptance of the machine. The beam at the target is well collimated, having a diameter of approximately 4 inches. The beam distribution on the target has a strong central maximum and is therefore very non-uniform under normal operating conditions.

The limitation on the maximum beam current available from A-48 is the power which can be dissipated on the target. The standard probe which is in use at present consists of a metal plate mounted in such a way that a high-pressure water flow of roughly 30 gallons per minute is maintained across the back. With copper targets it is possible to maintain heat loads of the order of 30 kw per square inch on  $\frac{1}{8}$ -in. copper plates. In the case of other materials, the heat load is usually smaller. Two wolfram targets were used in these experiments. One consisted of  $\frac{1}{16}$ -in. wolfram plate which was hard-soldered onto a  $\frac{1}{8}$ -in. copper plate. The other target was a  $\frac{1}{16}$ -in. wolfram plate mounted directly on the probe holder. Both of these targets were able to sustain heat loads of the order of 5 kw per square inch. The wolfram plate shattered completely toward the end of the second run. It is possible that the wolfram was

weakened (i.e., made brittle) by the large amount of hydrogen deposited on the plate during the run.

### II. ELECTRIC EXCITATION

The electric multipole (or Coulomb) excitation<sup>2</sup> of nuclei has been studied extensively during the last three years. Over one hundred energy levels in as many isotopes have been observed. The cross section for the excitation of most of the low-lying energy levels in the heavy nuclei is something of the order of several millibarns at the available proton energy. This process is particularly suited for the study of nuclear spectra because there are no unpleasant background radiations (i.e., neutrons) and it is therefore possible to observe the emitted  $\gamma$  rays very readily.

When a thick wolfram target is bombarded with 3.7-Mev protons, approximately 10<sup>-7</sup> nuclear  $\gamma$  rays are emitted per proton. The W targets which have been developed for the A-48 can be bombarded with proton beams up to 5 milliamperes. It is possible therefore to obtain equivalent  $\gamma$ -ray sources at the target of the order of 10<sup>10</sup>  $\gamma$  rays per second from electric excitation reactions. Such sources (~300 millicuries) are strong enough to make feasible the use of a bent-quartz-crystal spectrograph for the precision measurement of these  $\gamma$  rays.

### III. BENT-QUARTZ-CRYSTAL FOCUSING $\gamma$ -RAY SPECTROGRAPH

The bent-quartz-crystal focusing  $\gamma$ -ray spectrograph has been described in the literature.<sup>3</sup> The geometry of

\* California Institute of Technology, Pasadena, California.

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<sup>1</sup> E. O. Lawrence, *Science* **122**, 1127 (1955).

<sup>2</sup> Alder, Bohr, Huus, Mottelson, and Winther, *Revs. Modern Phys.* **28**, 432 (1956).

<sup>3</sup> Jesse W. M. DuMond, in *Ergebnisse der Exakten Naturwissenschaften* (Springer-Verlag, Berlin, 1955), Vol. 28, p. 232. (The