Electron Shakeoff Following the β^- Decay of Ar^{41}

THOMAS A. CARLSON
Oak Ridge National Laboratory, Oak Ridge, Tennessee
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A specially designed mass spectrometer is used to analyze the potassium ions that result from the β^- decay of Ar^{41}. The percent abundances are as follows: K^{1+} (82±1), K^{2+} (12.5±0.8), K^{3+} (3.0±0.2), K^{4+} (1.4±0.1), K^{5+} (0.44±0.06), K^{6+} (0.16±0.04), K^{7+} (0.06±0.02), and K^{8+} (0.016±0.009). The data are compared with similar studies on other rare gases that undergo beta decay, and some of the general properties of electron shakeoff as a function of atomic number are discussed.

I. INTRODUCTION

A SPECTRUM of positively charged potassium ions is formed as the result of the β^- decay of Ar^{41}. The singly charged ion is the consequence of the gain of one charge in the nucleus. Multiply charged ions are formed from the shakeoff of one or more orbital electrons. The relative abundances of the differently charged potassium ions that result from the β^- decay of Ar^{41} have been measured with a specially designed mass spectrometer and are presented in this paper.

The study on Ar^{41} completes a series of similar experiments on rare gases that undergo β^- decay. This series[1–5] includes all the rare gases in the Periodic Table from He to Xe. The purpose of these studies is to understand better the phenomenon of electron shakeoff that arises from the sudden change in nuclear charge. In this paper some of the general properties of electron shake off as a function of atomic number will be discussed.

II. EXPERIMENTAL

Ar^{41} was prepared from the reaction, $K^{41}(n,p)Ar^{41}$, by bombarding approximately 100 g of potassium aluminum silicate in the core of the Oak Ridge Research Reactor. The large surface area of the material permitted effective emanation; and the radioactive gas, which decays with a half-life of 110 min, was swept continuously by water vapor into the laboratory. An ice water trap removed most of the water vapor. Two more cold traps, a Cu-CuO oven, and a titanium pump helped purge the argon of contaminating gases.

The Ar^{41} was allowed to leak into a source volume of a mass spectrometer, previously described.[3,4,6] As the Ar^{41} underwent beta decay in the source volume, which contained a series of electrostatic field rings, the ions that were formed as the result of the decay were collected, focused, and then analyzed magnetically for their charge. The relative abundances of the differently charged K^{41} ions were compared under conditions of identical focusing by varying the collection voltage inversely to the charge.

The experiment was carried out with a total gas pressure of about 5×10^{-7} Torr in both the source volume and analyzer. Previous experience has shown us that at these pressures ion-molecule reactions do not affect the charge spectrum.[3] This was confirmed for the present study by observing no noticeable change in the charge spectrum at elevated pressures of about 1×10^{-5} Torr in the source volume.

The possibility was also checked that radiolysis of the other residual gases in the source volume might give rise to ions that could interfere with the results. The amount of radioactive gas was varied relative to the other gases. Also, the bulk gas was irradiated inside the source volume with electrons to determine what species would result. Neither of these tests indicated that there were any important contributions from radiolysis.

III. RESULTS AND DISCUSSION

The relative abundances of the differently charged K^{41} ions are given in Table I. The errors quoted are principally derived from counting statistics. It was noted in the study[2] on Ne^{23} that some of the singly charged ions apparently were not neutralized when they happened to strike the sides of the source volume, but, rather, were scattered off the walls. These scattered

TABLE I. Relative abundances of the potassium ions that result from the beta decay of argon (% abundance of the observed ion).

Charge of ion	Ar^{41}	Ar^{39} [a]
1	82 ±1	86 ±3
2	12.5 ±0.8	10 ±2
3	3.0 ±0.2	2.5 ±0.8
4	1.4 ±0.1	0.9 ±0.5
5	0.44 ±0.06	0.3 ±0.2
6	0.16 ±0.04	0.05±0.07
7	0.06 ±0.02	
8	0.016±0.009	

[a] See Ref. 9.

[1] He^6: T. A. Carlson, F. Pleasonton, and C. H. Johnson, Phys. Rev. **129**, 2220 (1963).
[2] Ne^{23}: T. A. Carlson, Phys. Rev. **130**, 2361 (1963).
[3] Kr^{85}: A. H. Snell and F. Pleasonton, Phys. Rev. **107**, 740 (1957).
[4] Xe^{133}: A. H. Snell and F. Pleasonton, Phys. Rev. **111**, 1338 (1958).
[5] T. A. Carlson, A. H. Snell, F. Pleasonton, and C. H. Johnson, in *Proceedings of a Symposium, Prague, 1960, on the Chemical Effects of Nuclear Transformations* (International Atomic Energy Agency, Vienna, 1961), Vol. I, p. 155.
[6] F. Pleasonton and A. H. Snell, Proc. Roy. Soc. (London) **A241**, 141 (1957).

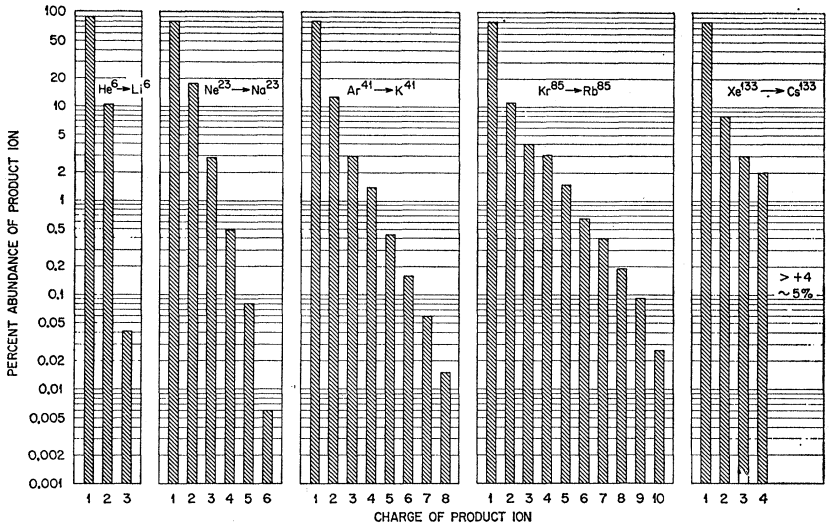

FIG. 1. Charge spectra of ions formed as a result of the β^- decay of He^6 (Ref. 1), Ne^{23} (Ref. 2), Ar^{41} (this paper), Kr^{85} (Ref. 3), and Xe^{133} (Ref. 4).

ions, which had been degraded in kinetic energy, were then collected with such high efficiency[7] that they caused an overestimation of the relative abundance of charge one. The contribution in this study to the relative abundance of K^{1+} from scattered ions has been estimated,[8] however, to be less than $\frac{1}{2}\%$. Also included in Table I are the relative abundances of the K^{39} ions that result from the β^- decay of Ar^{39}, as measured by Pleasonton.[9] Because of the low counting rates, and because of a rather high contamination of Ar^{40}, which necessitated radiolysis corrections, the results on Ar^{39} are rather inaccurate. Nevertheless, the two charge spectra in Table I are in reasonable agreement.

The data in Table I on the charge spectrum resulting from the decay of Ar^{41} are displayed graphically in Fig. 1, together with the results of similar studies on other rare gases. A number of papers have been written on the theoretical interpretation of electron shakeoff as the consequence of beta decay.[10–13] Specific calculations[1,2,14,15] have also been made on the probability of electron shakeoff during the decays of He^6, Ne^{23}, and Kr^{85}. Common to all of these papers is the concept that electron shakeoff is the consequence of the sudden nonadiabatic change in nuclear charge as the β^- particle leaves the atom with a velocity large compared to the velocity of the orbital electrons. The probabilities for electron shakeoff are obtained from the various transition probabilities to excited states in which the electrons are either in the continuum or in virtual states of the resulting ions. The transition probabilities may, in turn, be calculated from the squares of the overlap integrals between the initial state of the atom and the various states of the ion. The purpose of this discussion, however, is not to add any specific calculations but to restrict itself to a few general characteristics of electron shake off as revealed from the data on the rare gases.

From Fig. 1 it can be seen that the charge spectra follow rather regular patterns. It will be noted, for example, that for each spectrum (1) the singly charged ion is present in about 80% of the decays indicating that in most of the decays electrons are not shaken off, and (2) the remainder of the spectrum decreases monotonically with charge, the rate of decrease becoming smaller as the atom becomes more complex. It is of particular interest to compare the relative abundances of the doubly charged ions because they arise almost exclusively from the removal of one electron from the "outer shell."[16] Doubly charged ions are rarely observed as the consequence of the removal of an electron from one of the "inner shells"[16] because a vacancy thus created will lead, with nearly unit probability, to at least one self-ionization (Auger) process.[17] We see from the relative abundances of the charge-two ions (Fig. 1) that the amount of outer electron shakeoff is not strongly dependent on the atomic number. The probability for electron shakeoff has been estimated by several authors[11–13,15] who use the following general expression for any atom under-

[7] The collection efficiency of the spectrometer is roughly proportional to the kinetic energy of the ions being collected [T. A. Carlson, J. Chem. Phys. **32**, 1234 (1960)].

[8] The estimation has been made by assuming that the relative number of scattered ions and their kinetic energy are the same in both studies, so that the observed contributions of scattered ions will be directly proportional to the average recoil energy of the unscattered ions.

[9] F. Pleasonton (unpublished); see also Ref. 5.

[10] E. L. Feinberg, J. Phys. (U.S.S.R.) **4**, 423 (1941).

[11] A. Migdal, J. Phys. (U.S.S.R.) **4**, 449 (1941),

[12] J. S. Levinger, Phys. Rev. **90**, 11 (1953).

[13] H. M. Schwartz, J. Chem. Phys. **21**, 45 (1953).

[14] He^6: A. Winther, Kgl. Danske Viedenskab. Selskab. Mat. Fys. Medd. **27**, 2 (1952).

[15] Kr^{85}: A. E. S. Green, Phys. Rev. **107**, 1646 (1957).

[16] In this paper we shall regard the outer shell of a rare-gas atom as made up of the outermost s and p states. All the other electrons in the atom will be defined as inner shell electrons.

[17] See, for example, the arguments in Ref. 3. Also see the Auger transition probabilities as calculated by R. A. Rubinstein, Doctoral thesis, University of Illinois, 1955 (unpublished).

TABLE II. Comparison of $A_2(\bar{Z}_{\text{eff}})^2$ with $(\alpha_{ns}+\alpha_{np})$ for rare gases undergoing β^- decay.

Parent atom	Z	n	$A_2(\%)$[a]	$\bar{Z}_{\text{eff}}$[b]	$A_2(\bar{Z}_{\text{eff}})^2$	$(\alpha_{ns}+\alpha_{np})$ [c]	[d]	[e]
Ne^{23}	10	2	17.5	5.9	6	4.7	2.3	6.8
Ar^{41}	18	3	12.5	8.7	9	7.6	4.3	14
Kr^{85}	36	4	10.9	13.4	20	19.5		
Xe^{133}	54	5	8	18.7	28			

[a] Relative abundance of the doubly charged ion from data in Table I and Refs. 2, 3, and 4.
[b] See Ref. 18.
[c] A. E. S. Green, Refs. 15 and 19.
[d] H. M. Schwartz, Ref. 13.
[e] A. Migdal, Ref. 11.

going β^- decay:

$$P_{nl}=\alpha_{nl}/(Z_{\text{eff}})^2, \tag{1}$$

where P_{nl} is the probability for shaking off an electron from the nl shell (n is the principal quantum number and l is the angular momentum quantum number). α_{nl} is a constant for the nl shell and is assumed to hold for any atom. Z_{eff} is the effective charge as seen by the electron to be removed. From Eq. (1) it follows that

$$[A_2(\bar{Z}_{\text{eff}})^2]_n \approx \alpha_{ns}+\alpha_{np}, \tag{2}$$

where A_2 is the experimentally determined relative abundance of the doubly charged ion resulting from the decay of a rare-gas atom whose outer shell has the principal quantum number, n, and $\bar{Z}_{\text{eff}}$ is the average effective charge[18] as seen by the s and p electrons of the outer shell. In Table II are listed $A_2(\bar{Z}_{\text{eff}})^2$ for each of the rare gases from Ne to Xe. [The He^6 study is not included for comparison because (1) the general theory does not apply to very light elements, and (2) transitions to virtual states are of major importance in He^6 [1] while of minor importance for the other rare gases.[2,15]] It appears from Table II that $[A_2(\bar{Z}_{\text{eff}})^2]_n$ increases for larger values of the principal number. The theoretically obtained values of $\alpha_{ns}+\alpha_{np}$ are also given in Table II from the work of several authors.[11,13,19]

[18] The average effective charges as listed in Table II have been somewhat arbitrarily defined as $[Z_{\text{eff}}(s)+Z_{\text{eff}}(p)]/2$, where $Z_{\text{eff}}(s)$ and $Z_{\text{eff}}(p)$ are, respectively, the effective charges seen by the outer s and p electrons of the rare-gas atom in question. $Z_{\text{eff}}(s)$ and $Z_{\text{eff}}(p)$ were obtained, with the exception of the values for Xe, from screening constants given by Froese as a function of the mean radii [C. Froese, Proc. Roy. Soc. (London) **A239**, 390 (1958)]. The mean radii for the $4s$ and $4p$ states of Kr, which were not given in the above references, were calculated from radial wave functions of Kr [B. H. Worsley, Proc. Roy. Soc. (London) **A247**, 390 (1958)]. The screening constants for Xe were obtained from calculations on Cs^+ as given by Ridley [E. C. Ridley, Proc. Cambridge Phil. Soc. **51**, 693 (1955)].

[19] For Green's calculations we have used the relationship,

$$\alpha_{nl}=\left[\bar{Z}^2 P_v \cdot 2(2l+1)P \cdot \frac{1}{P_v}\right]_{nl},$$

where the symbols are defined in Ref. 15; their values are given on p. 1649 of the same reference.

The agreement between theory and experiment seems, in general, quite satisfactory.

The formation of ions with charges greater than two may come about in two ways: (1) multiple electron shakeoff of the outer shell electrons and (2) shakeoff of inner orbital electrons, followed by Auger processes. Multiply charged ions in the β^- decay of He^6 and Ne^{23} have been discussed primarily in terms of the first process.[1,2] However, since the probability for removal of one electron from the outer shell (and presumably the removal of more than one electron from this shell) does not increase as one goes to heavier rare gases, it seems that the observed increase in the complexity of the spectra for the heavier atoms must be attributed to the greater number of inner orbital electrons that can be shaken off. Also as the atom becomes larger, the removal of a K or L electron can give rise to an extensive number of Auger processes that result in the production of highly charged ions.[20] The Z_{eff}, however, will also increase for a given shell as the atomic number increases, which will cause the probability for shaking off an electron in that shell to decrease. Thus, we find in Fig. 1 that as we go to the heavier rare gases the spectra become more complex, including a greater variety of ions of increasingly higher charges, but the total number of multiply charged ions does not increase appreciably.

IV. CONCLUSION

The charge spectra of the alkali metal ions that result from the β^- decay of the rare gases reveal a rather coherent picture. The spectra all show a monotonic decrease in intensity with increase in charge. The probability for not observing any electron shakeoff, as given by the abundances of the singly charged ions, remains fairly constant as a function of atomic number. The probability for shaking off an outer shell electron, as given by the doubly charged ions, can be satisfactorily correlated with theoretical predictions. The principal change in the spectra as one goes to more complex atoms is the increase in the relative number of the multiply charged ions, which is interpreted as an increase in the importance of the removal of inner shell electrons.

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[20] A. H. Snell, F. Pleasonton, and T. A. Carlson, in *Proceedings of a Symposium, Prague, 1960, on the Chemical Effects of Nuclear Transformations* (International Atomic Energy Agency, Vienna, 1961), Vol. I, p. 147.