Charge Distribution for ^{133}Xe - ^{133}Cs Decay Process

KAZUSHIGE NISHIZAWA*, KAORU NARISADA*, HIDENORI TERAMATSU*,
MASATAKA SOYA* AND MUTSUAKI SHINAGAWA*

(Received 31 January 1973)

Charge distributions of cesium ions following beta and internal conversion decays of ^{133}Xe were investigated both theoretically and experimentally. The calculations were carried out on the assumption that reorganization of the electronic configuration occurred at each successive step in a vacancy-cascade in which Auger and radiative transitions were involved. The electron-shakeoff process following negative beta decay was also incorporated in the calculation. The calculated spectrum was compared favorably with the experimental one which had been measured with a charge spectrometer.

It was found that the vacancy-cascade model, which consisted of radiative, Auger, and shakeoff processes, was satisfactory to account for the charge distribution of an atom followed by a nuclear decay.

1. Introduction

The inner shell vacancy produced by a nuclear decay will be filled by the transition of the outer shell electron with the emission of another electron (Auger electron) or alternatively of characteristic X-ray (fluorescence X-ray). As the results of nonradiative and radiative transitions, an original vacancy will lead outer shells to the numerous holes. The reorganization of an atom with the stepwise processes of Auger and radiative transitions was first recognized as a vacancy-cascade by Cooper.¹⁾ If the vacancy-cascade occurs at the high Z atom, this atom will be multiply ionized. As it is well known, an initial vacancy is created by the nuclear decay, like electron capture or internal conversion. The inner shell ionization is also caused by the interaction of the shell electron with the external radiation, as in the case of photoelectric ionization.

This process of reorganization of an atom has been used by several authors to compute

successfully charge distributions that result from electron capture^{2), 3)} in ^{37}Ar , and from K photoionization⁴⁾ of argon. Charge distribution⁵⁾ of krypton following ionization in the 1s, 2s or 2p shell was calculated thereafter by Krause and Carlson on the assumption that reorganizations occurred by successive steps in a vacancy-cascade in which Auger and radiative transitions partook. These distributions were compared favorably with experimental charge spectra arising from photo-ionization in the K and L shell, from electron capture in ^{79}Kr and from β decay in ^{85}Kr .

Xenon-133 disintegrates to the ground state of ^{133}Cs in the mode as shown in Fig. 1. In this decay process, the orbital electrons of the daughter atom suffer the rapid change of the electric field as a result of the sudden change in the nuclear charge, so some of them are shaken off to the continuum, and then one of the inner shell electrons may be emitted through the internal conversion.

Relative transition rates⁶⁻¹²⁾ of both

* Faculty of Engineering, Osaka University (Yamadakami, Suita-shi, Osaka)

Fig. 1. The decay scheme of ^{133}Xe to ^{133}Cs . The branch that is heavily indicated dominates greatly in intensity. The 81 keV level decays through the internal conversion of 68.2 %. Transcribed from Lederer.¹⁴⁾

radiative and nonradiative processes have been recently published on some series of shells except on N to O shell transition. Probabilities of electron-shakeoff¹³⁾ following β -decay are known on $Z=55$ and internal conversion rates¹⁴⁾ to 1s, 2s and 2p are sufficiently well known on ^{133}Cs in excited states. Values of these parameters entering into the model of the calculation are available from independent experiments or from theoretical predictions, most of which have proven reliable by different types of experiments.

In the following section we present individual charge distributions following the electron-shakeoff, internal conversion, and vacancy-cascade for the sake of basic considerations to try further calculation with Monte Carlo Method to obtain a theoretical spectrum. To compare with it, we present also the experimental spectrum of ^{133}Cs obtained with a charge spectrometer.

2. Basic Considerations for Calculation

2.1 Electron-Shakeoff

The nuclear charge changes from Z to $Z+1$, as the result of negative beta decay.

The orbital electrons find themselves in the different electric field, and some of them are released from the shell. The electron-shakeoff takes place when the electric field rapidly changes not only in the negative beta decay, but also in the case of photoelectric ionization or of the Auger transition. To determine the charge distribution following negative beta decay, we use the shakeoff probabilities that have been calculated by Carlson.¹³⁾ As the electron-shakeoff is more predominant in the outer shell than in the inner shell, the vacancy-cascade initiated from negative beta decay is less probable than from photo-ionization or internal conversion. Table 1 shows the charge spectrum of ^{133}Cs as a result of the shakeoff followed by the beta decay of ^{133}Xe and it does not involve the result of the internal conversion.

2.2 Internal Conversion

As it is seen in Fig. 1, the strong branch to the level 81 keV of ^{133}Cs is noticed on the occasion of ^{133}Xe beta emission. It decays to the ground state with radiative emission of which 68.2 percent is due to the internal conversion. This internal conversion is of the

Charge Distribution for ^{133}Xe - ^{133}Cs Decay Process

Table 1. Charge spectrum of ^{133}Cs calculated with shakeoff probabilities⁽¹⁾ following β^- -decay of ^{133}Xe .

Charge	(%)
0	25.67
1	3.79
2	1.65
3	0.23
4	0.17
5	0.03
6	0.10
7	0.01
8	0.04
9	0.02
10	0.01
Total	31.72

intense decay mode that accompanies the vacancy-cascade particularly, for it produces the vacancy in an inner shell of the cesium atom. The probability of the internal conversion to each shell shown in Table 2 represents the rate of produced vacancies per beta emission.

Table 2. Internal conversion coefficients⁽⁴⁾ of the level 81 keV to 1s, 2s and 2p electrons, arranged in rates of produced vacancies per β^- -decay of ^{133}Xe - ^{133}Cs .

1s	0.583
2s	0.077
2p	0.022
Total	0.682

2.3 Vacancy-Cascade

A hole produced in an inner shell is filled by the radiative transition with the emission of a quantum or, alternatively, by the Auger transition with the emission of an electron. We will trace all possible transition routes leading from the initial vacancy to the outer shells and note for each path the number of electrons lost in Auger events. Then we will be able to find out each intensity for every route from the relative rates of the individual transitions, which occur successively one

after the other within a route. Finally, we obtain the general charge distribution by grouping together paths that lead to the same charge states.

3. Calculation with Monte Carlo Method

To calculate individual charge distributions that follow after the removal of a 1s, 2s or 2p electron in xenon, we simulated the above mentioned model of the vacancy cascade by a computer program. For this purpose, we employed a Monte Carlo method using FORTRAN programs for the NEAC 2200-700 computer. The computer picked up stepwise all possible transitions concerning a given vacancy or vacancies according to their relative rates. After 50,000 cases had been computed, the computer registered the final number of vacancies, namely the charge distributions. For the computation, we made use of the transition rates listed in Table 3, which were of radiative and Auger transitions in a singly ionized atom. The flow diagram of the calculation was, as shown in Fig. 2, for the transitions of M-shell vacancies. If a vacancy was created in the 3d-shell by a transition from an inner shell or by an electron shakeoff, it must be propagated in N- and/or O-shell. The routes of these possible transitions were determined by the difference between the transition probability P_n and the random number RN_i which was produced by the sub-routine program of the computer.

The charge spectra calculated with the cascade model are shown in Table 4; the 2nd, 3rd and 4th columns of which are the final charge distributions which have been derived mainly from the internal conversions and the 5th, 6th and 7th columns are those from the electron-shakeoffs.

Keeping the transition rates per electron being constant, we ignored the changes of transition rates due to changes of the atomic

Table 3. Relative transition rates for radiative and Auger transitions in cesium.

Initial state	Final state	Transition rate	Initial state	Final state	Transition rate	Initial state	Final state	Transition rate
1s	2p ¹⁰¹	76.3	2p	3d ¹⁰¹	9.4	3p ⁷¹	3p5p	1.0
	3p ¹⁰¹	13.9		4d ¹⁰¹	1.5		3d4s	14.3
	4p ¹⁰¹	2.9		2p4s ¹¹¹	0.6		3d4p	55.5
	2s 2s ⁹¹	0.6		2p4p ¹¹¹	3.5		3d4d	12.5
	2s 2p ⁹¹	1.7		2p4d ¹¹¹	5.3		3d5s	1.9
	2p2p ⁹¹	3.1		2p5p ¹¹¹	0.5		3d5p	5.8
	2p3s ⁹¹	0.5		3s 3p ¹¹¹	2.7		4s 4p	0.6
	2p3p ⁹¹	1.0		3s 3d ¹¹¹	0.6		4s 4d	0.6
2s				3p3p ¹¹¹	10.9		4p4p	2.4
	3p ¹⁰¹	4.2		3p3d ¹¹¹	19.7		4p4d	5.4
	4p ¹⁰¹	1.0		3p4p ¹¹¹	3.1	3d ⁷¹	4s 4d	4.9
	2p4s ¹¹¹	10.3		3p4d ¹¹¹	2.7		4p4p	1.8
	2p4p ¹¹¹	17.9		3d3d ¹¹¹	30.0		4p4d	24.8
	2p4d ¹¹¹	18.9		3d4p ¹¹¹	1.9		4d4d	61.6
	2p5s ¹¹¹	1.7		3d4d ¹¹¹	7.6		4d5s	2.2
	2p5p ¹¹¹	2.6					4d5p	4.7
	3s 3s ¹¹¹	1.2	3s ⁷¹	3p4p	28.4			
	3s 3p ¹¹¹	7.4		3p4d	32.9			
	3s 3d ¹¹¹	10.3		3p5s	1.0			
	3s 4p ¹¹¹	1.0		3p5p	2.6			
	3s 4d ¹¹¹	1.5		3d4s	9.8			
	3p3d ¹¹¹	5.4		3d4p	12.0			
	3p4s ¹¹¹	0.7		3d4d	6.0			
	3d3d ¹¹¹	12.3		4s 4d	1.9			
	3d4s ¹¹¹	1.0		4p5s	1.5			
	3d4d ¹¹¹	2.6		4p5p	4.1			

field whenever an additional inner-shell electron had been removed. This introduced only a slight error since an Auger transition was by far the most likely events in the reorganization process and its rate was usually never perturbed by the change of the field.

In the transitions from N-shell to O-shell, we only restricted ourselves to the N-OO processes, regardless of the Coster-Kronig transitions and the double Auger effects, N-OOO, both of which seemed to take place probably. Such transition as the Coster-Kronig or double Auger, should have been thought to lead to an odd number for the final

charge, while the N-OO transition to an even charge.

4. Experimental

4.1 Apparatus

The apparatus employed for these measurements was the charge spectrometer which we constructed as has been reported previously,^{15), 16)} and its mode of use has been essentially unchanged. Since the charge spectrometer had different collection efficiencies for differently charged ions, we corrected the measured values by applying the charge discrimination factors which had been obtained with the

Fig. 2. Flow diagram of the Monte Carlo method for M-shell vacancies.

P_n : the transition rate listed in Table 3.

RN_i : the random number produced by the subroutine program of NEAC 2200-700.

3s, 3p, 3d, N and O: representation of the corresponding shell.

Table 4. Individual charge distributions of ^{133}Cs ions following an initial vacancy of each shell. Calculations were made according to the vacancy-cascade model.

Charge	1s	2s	2p	3s	3p	3d
1	0	0	0	0	0	0
2	3.97	0.938	1.53	0	0	0
3	0.476	0	0.632	5.60	0	6.90
4	7.98	0.790	9.08	1.90	9.5	93.1
5	2.44	1.91	0.768	2.20	12.9	0
6	18.6	11.6	10.4	32.2	76.8	0
7	4.09	4.26	4.87	10.7	0.8	
8	28.8	25.4	38.0	46.9	0	
9	18.2	32.2	22.9	0.50	0	
10	9.13	16.8	10.9			
11	2.13	5.52	1.06			
12	1.43	0.552	0			
13	1.41	0	0			
14	0.712	0				
15	0.448					
16	0.104					
17	0.120					
18	0.014					
19	0.012					

measurement of the charge distribution of ^{85}Kr - ^{85}Rb .

4.2 Experimental Procedure

The quantity of the radioactive gas in the source volume, where the ions were yielded, was kept constant during the measurement by adjusting the leak valve depending on the ionization gauge and the monitor counter (a NaI (TI) scintillation counter). The total pressure in the source volume was 1×10^{-6} Torr and it was increased to 2×10^{-5} Torr after the introduction of xenon.

The ions produced in the source volume were collected by the guide electrodes supplied 350 V across the first to 15th, and they were emitted from the apex of the source volume with energy of 4350 eV per unit charge.

Radioactive xenon, ^{133}Xe , was purchased from the Radiochemical Center (England). Our main reliance on purity was based upon the specification. It had been described that the radiochemical purity was 99 percent or more, and the specific activity was above 10 Ci/cm³ at S.T.P.

4.3 Experimental Results

The results of the ^{133}Xe - ^{133}Cs measurements are given in Table 5. They are indicated as white columns in the semilogarithmic histogram of Fig. 3, in comparison with the shaded columns of calculated values. The counting rate for each peak of the original charge spectrogram was obtained by the detailed sweep over the corresponding channel of the multiscaler. Determining the channels corresponding the peaks of $^{85}\text{Rb}^+$ and $^{87}\text{Rb}^+$ produced by surface ionization, we calculated the intensity of the magnetic field and the corresponding peak channel for each charge. The data of ^{133}Cs given in Table 5 are the corrected values as for the collecting efficiencies obtained with ^{85}Rb and ^{87}Rb and are the average of thrice measurements. For the

Table 5. Experimental charge spectrum resulting from beta and internal conversion decays of ^{133}Xe .

Charge of ions	Intensity(%)	Charge of ions	Intensity(%)
1	32.1	11	7.82
2	3.80	12	2.89
3	2.26	13	1.06
4	1.51	14	0.99
5	2.01		
6	3.51		
7	6.25		
8	9.40		
9	13.1		
10	13.3		

weak specific activity of ^{133}Xe obtained and for the smallness of the source volume 20 liters, we could not detect above 15 of charges, while the calculation indicated the existence of ions

up to charge 20.

5. Comparison of Experimental Results with Calculated Charge Spectra

In Fig. 3 the experimental charge distribution, arising from the nuclear decay of ^{133}Xe , is compared with the calculated spectrum, i.e. the calculated results by the stepwise reorganization model of the atom. Since they indicate the fairly good agreement, we see that the vacancy-cascade model which incorporates radiative, Auger and shakeoff processes is capable of accounting satisfactorily for the experimental charge distribution. Auger events are responsible for the bulk of ionization and the general shape of the distribution; the electron-shakeoff smoothes the spectrum out and shifts it toward the higher charge end.

Fig. 3. Comparison of experimental and calculated charge spectra resulting from ^{133}Xe - ^{133}Cs decay.

The discrepancies at the lower charges are caused by fault of the assumption taken for calculation. Because of the insufficiency of the data in literatures, we only adopted the N-OO transition as for a N-shell vacancy, and disregarded the Coster-Kronig transition and the double Auger event that might lead to release two electrons at the outermost shell. These discrepancies caused by the simplification show that the probabilities of Coster-Kronig and double Auger events should be considerably larger in the N- to O-shell transitions.

When we examine the high charge slope of the spectra shown in Fig. 3, an unexpected feature emerges. The calculated values are generally smaller than the experimental ones. In this respect, one possible reason comes from the neglect of the electron-shakeoff following the rapid change of the electrostatic field caused by the electron-ejection, i.e. Auger, shakeoff, or internal conversion. Otherwise it can be ascribed to the N-OO transition as has been mentioned above.

6. Conclusion

The experimental charge spectrum of ^{133}Xe - ^{133}Cs almost agrees with the calculated spectrum on the assumption of simple N-OO transition. Though the N-OO transition has to be recognized as the leading mechanism, the small

discrepancies suggest the necessity of additional considerations. It must be emphasized that the complete charge state should be computed with the transition rates in multifold ions. The authors hope further development in this respect.

Acknowledgment

The authors greatly appreciate the valuable help of Mr. T. Yamagishi for setting up the Monte Carlo program.

References

- 1) E. P. Cooper, *Phys. Rev.*, **61**, 1 (1942).
- 2) R. A. Rubenstein and J. N. Snyder, *Phys. Rev.*, **99**, 189 (1955).
- 3) A. H. Snell and F. Pleasonton, *Phys. Rev.*, **100**, 1396 (1955).
- 4) T. A. Carlson and M. O. Krause, *Phys. Rev.*, **137**, A1655 (1965).
- 5) M. O. Krause and T. A. Carlson, *Phys. Rev.*, **158**, A18 (1967).
- 6) E. J. McGuire, *Phys. Rev.*, **A5**, 1043 (1972).
- 7) E. J. McGuire, *Phys. Rev.*, **A5**, 1052 (1972).
- 8) B. Crasemann, M. H. Chen and V. O. Kostroun, *Phys. Rev.*, **A4**, 2161 (1971).
- 9) P. V. Rao, M. H. Chen and B. Crasemann, *Phys. Rev.*, **A5**, 997 (1972).
- 10) J. H. Scofield, *Phys. Rev.*, **179**, 9 (1969).
- 11) E. J. McGuire, *Phys. Rev.*, **A3**, 1801 (1971).
- 12) E. J. McGuire, *Phys. Rev.*, **A3**, 587 (1971).
- 13) T. A. Carlson, C. W. Nestor, T. C. Tucker and F. B. Malik, *Phys. Rev.*, **169**, 27 (1968).
- 14) C. M. Lederer, *et al.*, "Table of Isotopes", Sixth Edition, John Wiley and Sons, New York (1967).
- 15) K. Nishizawa, K. Narisada and M. Shinagawa, *Mass Spectroscopy*, **17**, 854 (1969).
- 16) K. Nishizawa, K. Narisada, H. Teramatsu and M. Shinagawa, *Mass Spectroscopy*, **18**, 1149 (1970).