

Electron Shake-Off Following the Beta Decay of Ne^{23}

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A specially designed mass spectrometer is used to analyze the sodium ions that result from the β^- decay of Ne^{23} . The percent abundances are as follows: Na^{1+} (79.1), Na^{2+} (17.5), Na^{3+} (2.85), Na^{4+} (0.48), Na^{5+} (0.081), Na^{6+} (0.006), Na^{7+} (<0.002). No evidence is found that recoil energy contributes to the electron shake-off. An attempt is made to derive the charge spectrum from the assumption that electron shake-off is due to the sudden, nonadiabatic change in nuclear charge. Multiple electron shake-off is computed from the product of the probabilities for single electron shake-off. The probabilities for single electron shake-off are semiempirically calculated. In general, the calculated values agree well with the experimental data, though the computations underestimate the abundances of the more highly charged ions.

I. INTRODUCTION

A SINGLY charged ion is formed following β^- decay because of the gain of one charge in the nucleus. Further ionization may occur because of excitation that arises from the sudden change in nuclear charge. The extent of this ionization has been measured for several rare gases that undergo β^- decay.¹⁻⁴ In these experiments the rare gases were allowed to decay in a specially designed mass spectrometer, and the relative abundances of the ions formed were measured. This paper contains the experimental results for the charge spectrum of the sodium ions arising from the β^- decay of Ne^{23} . Data are also included for setting a limit to the amount of electron shake-off in the decay of Ne^{23} that might result from recoil energy.

Because of the simple structure of helium, Winther⁵ was able to calculate with some precision the probability for single electron shake-off following the β^- decay of He^6 . The excellent agreement between Winther's calculation and experimental data³ has placed the theory for single electron shake-off on a firm basis. For an understanding of the multiple shake-off process, however, one must go to a more complex atom. The next heavier rare gas, neon, is a logical choice for study, and it can be readily produced in a form that undergoes beta decay, namely, Ne^{23} .

This paper also contains a calculation of the charge spectrum for the ions formed from the decay of Ne^{23} . First, the probabilities are calculated for single electron shake-off. Then the extent of multiple electron shake-off is obtained from the products of the probabilities for single electron removal. Finally, the charge spectrum is evaluated from the amount of multiple electron shake-off, after small corrections are made for Auger processes.

¹ Kr^{85} : A. H. Snell and F. Pleasonton, *Phys. Rev.* **107**, 790 (1957).

² Xe^{138} : A. H. Snell and F. Pleasonton, *Phys. Rev.* **111**, 1338 (1958).

³ He^6 : T. A. Carlson, F. Pleasonton, and C. H. Johnson, *Phys. Rev.* **130**, 2220 (1963).

⁴ Ar^{41} : T. A. Carlson, Oak Ridge National Laboratory, *Phys. Div. Ann. Progr. Rept.* 1962, ORNL-3268, p. 65 (unpublished).

⁵ A. Winther, *Kgl. Danske Videnskab. Selskab, Mat. Fys. Medd.* **27**, No. 2 (1952).

II. EXPERIMENTAL

Ne^{23} was prepared from the reaction, $\text{Na}^{23}(n,p)\text{Ne}^{23}$, by bombarding approximately 100 g of sodium 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 40 sec, was swept continuously by water vapor into the laboratory. An ice water trap removed most of the water vapor. Two more cold traps and a Cu-CuO oven helped purge the neon of contaminating gases.

The charge spectrum of Na^{23} ions that results from the β^- decay of Ne^{23} was measured with a specially designed mass spectrometer, which has previously been described in some detail^{1-3,6,7}. Ne^{23} was allowed to decay in a source volume. A portion of the Na^{23} ions that were formed from the decay were extracted. These ions were then analyzed with a tandem arrangement of magnetic and electrostatic analyzers.

Two different source volumes were used in the present experiment. One is a field-free source volume, which was designed by Snell and Pleasonton for their work on⁶ Ar^{37} . Its conical shape allows the recoil Na^{23} ions to emerge as a beam. The principal advantage of such a system is that it offers one a chance to measure the recoil energy as well as the charge of the ions. The recoil energy spectrum from the decay of Ne^{23} , in fact, will be a subject for a future publication.⁸ The principal disadvantage is that only about 10^{-6} of the ions emerge from the cone. The second source volume used was designed for the purpose of enhancing the collection efficiency.⁷ The ions are collected and focused by a series of electrodes in the shape of conical sections. On emerging from the source volume, the ions are accelerated before entering the spectrometer so as to decrease their relative energy spread.

Regardless of which source volume was employed or whether preacceleration or postacceleration was used

⁶ A. H. Snell and F. Pleasonton, *Phys. Rev.* **100**, 1396 (1955).

⁷ F. Pleasonton and A. H. Snell, *Proc. Roy. Soc. (London)* **A241**, 141 (1957).

⁸ T. A. Carlson (to be published). See also Oak Ridge National Laboratory, *Phys. Div. Ann. Progr. Rept.* 1962, ORNL-3268, p. 62 (unpublished).

during the analysis, all field strengths in a given run were varied proportionally to the charge of the analyzed ions, so that the relative abundances of the differently charged ions were compared under conditions of identical focusing and equal final energies.

The analyses were carried out with total gas pressures of about 2 to 5×10^{-6} Torr in the source volumes and about 1×10^{-6} Torr in the analyzers. Previous experience has shown us that at these pressures ion-molecule reactions do not affect the charge spectrum.¹ This was confirmed for the present study by observing no noticeable change in the charge spectrum at elevated pressures of about 1 to 2×10^{-5} Torr in the source volumes.

The possibility was also checked that radiolysis of the bulk gas might result in ions that could interfere with the analysis of the sodium ions. The amount of radioactive gas was varied relative to the bulk gas. 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 contributions from radiolysis.

III. RESULTS

A. Charge Spectrum

The relative abundances of the differently charged sodium ions are compared at the same recoil energy. The choice of this recoil energy is arbitrary, since recoil energy in the decay of Ne^{23} plays a negligible role in shaking off electrons (see Sec. III B). Most of the data taken with the field-free source volume were for ions having a recoil energy of 370 eV. When the ion-collection source volume was used, the spectrometer was set to receive ions that had nearly zero recoil energy, since this source volume collects the lower energy ions most efficiently. The results for the relative abundances of the differently charged ions, as obtained from the two different source volumes, are given in Table I. With one exception, which will be discussed below, the errors quoted are those from counting statistics. Although there is a slight hint that the analyses as given for the field-free source volume may favor the more highly charged ions as compared to the ion-collection source volume, the agreement between the two sets of data is generally good, and promotes confidence in the experimental procedures. The charge spectrum of the Na^{23} ions

is obtained from the results in Table I. When values are available from both source volumes, their weighted average is used.

One problem, however, should be discussed. Though not designed for the task, the ion-collection source volume can be used to measure the charge spectrum of the sodium ions as a function of the recoil energy. As expected, no dependence on recoil energy was observed for the relative abundances of sodium ions having a charge greater than one; but, unexpectedly, the charge-one ion did seem to be dependent on recoil energy. In Fig. 1 the ratio of ($\text{Na}^{2+}/\text{Na}^{1+}$) is plotted against the recoil energy of the ions. The ratio of ($\text{Na}^{2+}/\text{Na}^{1+}$) at first rises quickly as the recoil energy increases, but then levels out at higher energies to a value in reasonable agreement with the data obtained from the field-free source volume. This behavior is far too drastic to be accounted for by electron shake-off. Also the effect,

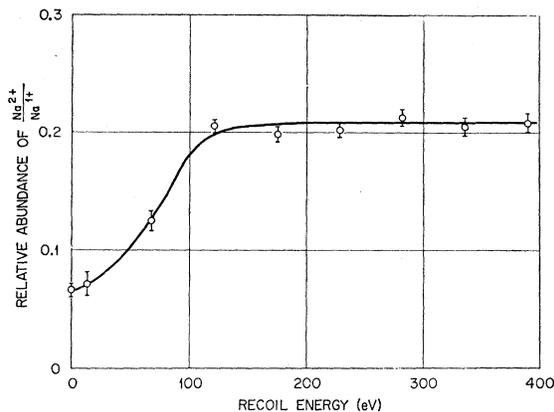


FIG. 1. Dependence on recoil energy of the relative abundance of singly to doubly charged Na ions formed from the decay of Ne^{23} . (Data taken with an ion-collection source volume under conditions where the resolution of the spectrometer is $\sim \pm 50$ eV at half-maximum.)

rather than being linearly dependent on the recoil energy, is present only at low recoil energies. The data in Fig. 1 can be better understood when we consider that, unlike the field-free source volume, the ion-collection source volume contains no baffles to prevent the inclusion of scattered ions. Normally this has not been a problem since the ions are rendered neutral when they strike the walls of the source volume. But two facts cause the appearance of this problem in the present study. First, the electron affinity for the singly charged sodium ion is 5.1 eV, which is rather small when compared with most positive ions. Second, the collection efficiency for an ion that has a small kinetic energy may be several orders of magnitude greater than that for the average Na^{23} ion, whose recoil energy is about 250 eV. Thus, even if only a few Na^{1+} ions are degraded in energy by scattering from the walls, they might form a sizable contribution to the total number of ions observed. In previous studies the problem of scattered

TABLE I. Comparison of data obtained from the field-free source volume with those from the ion-collection source volume.

Ions compared	Relative abundances of ions	
	Field-free source volume	Ion-collection source volume
$\text{Na}^{2+}/\text{Na}^{1+}$	0.2214 ± 0.0012	0.208 ± 0.008
$\text{Na}^{3+}/\text{Na}^{2+}$	0.165 ± 0.004	0.160 ± 0.006
$\text{Na}^{4+}/\text{Na}^{2+}$	0.0275 ± 0.0015	0.0278 ± 0.0018
$\text{Na}^{5+}/\text{Na}^{2+}$	0.0054 ± 0.0007	0.0043 ± 0.0005
$\text{Na}^{6+}/\text{Na}^{2+}$	0.001 ± 0.002	0.00036 ± 0.00013
$\text{Na}^{7+}/\text{Na}^{2+}$		0.00005 ± 0.00005

ions has not proved to be of significant importance.⁹ In one sense, the appearance of scattered Na^{23} ions has been the exception that proves the rule, since only under the most disadvantageous circumstances has the contamination of scattered ions been observed. From the above arguments, the "true" value of $\text{Na}^{2+}/\text{Na}^{1+}$, as obtained from the ion-collection source volume, is the ratio taken at high recoil energy. This is the value listed in Table I. The error quoted for the ratio is based more on the ability to set the spectrometer accurately than on the counting statistics. The sensitivity of this setting comes from the fact that the initial recoil energy is only a small fraction of the total energy of the ions analyzed.

B. Effect of Recoil Energy on Electron Shake-Off

If recoil energy is capable of inducing extensive electron shake-off, one would expect to observe a linear increase in the relative abundances of the more highly charged ions, as the recoil energy increases.¹⁰ As discussed in the previous section, data with the ion-collection source volume did not show this effect. It seemed worthwhile, however, to obtain more accurate data on the effect of recoil energy since a small contribution to electron shake-off due to recoil energy was noted in the decay of He^6 .³ In Table II are listed various relative abundances of sodium ions taken at two different recoil energies with the field-free source volume. Within the error of counting statistics no recoil effect is noted. This is to be expected, since the maximum recoil energy received by Li^6 in the decay of He^6 is nearly three times that received by the sodium ions in the decay of Ne^{23} .

IV. DISCUSSIONS AND CALCULATIONS

Calculation of the probability for shaking off an electron during β^- decay has been discussed by a number of authors.^{5,11-15} During β^- decay there is a sudden change in the nuclear charge. The electrons of the decaying atom can become excited because the β^- particle leaves too quickly for the orbital electrons to adjust adiabatically to their new environment. This excitation may be evaluated by the sudden approximation¹⁶ if the time spent by the β^- particle in traversing an electron shell is short compared to the period of revolution for the electron in that shell. This will be true for a relativistic β^- particle if $Z_{\text{eff}}\alpha$ is small with

TABLE II. Dependence of the relative abundance (%) of Na ions formed from the decay of Ne^{23} on recoil energy (data taken with field-free source volume).

Ions compared	Relative abundance at 250 eV	Relative abundance at 450 eV
2/1	0.219 \pm 0.006	0.220 \pm 0.005
3/2	0.173 \pm 0.007	0.167 \pm 0.006
4/2	0.029 \pm 0.003	0.026 \pm 0.002
5/2	0.005 \pm 0.001	0.006 \pm 0.001

respect to unity, where Z_{eff} is the effective charge and α is the fine structure constant. The sudden approximation should hold quite well for the decay of Ne^{23} . To determine the transition probability to a final state, one squares the amplitude for the transition as given by the overlap integral of the ground state of the initial atom and the final state of the resultant ion.

A. Single Electron Shake-Off

The transition probability for a single electron to go from an nl state in Ne^0 to an $n'l'$ state in Na^+ is given by

$$P(nl, \text{Ne}^0 \rightarrow n'l', \text{Na}^+) = \left| \int \psi_{n'l', \text{Na}^+}^* \psi_{nl, \text{Ne}^0} d\tau \right|^2 \quad (1)$$

where n and n' are the principal quantum numbers, l and l' are the orbital angular momentum quantum numbers, and $l-l'=0$. In this paper we shall treat the transition probabilities for each electron as independent of the other electrons. Also, each electron of Ne^0 or Na^+ , as the case may be, that has the same principal and orbital angular momentum quantum numbers will be described by the same wave function.

One would like to calculate from Eq. (1) the probabilities that electrons initially in $1s$, $2s$, or $2p$ states of neon will find themselves, following β^- decay, in some state that will undergo ionization. In order to enter into such a state the electron must vacate its initial state. We first shall calculate $P(nl, \text{Ne}^0 \rightarrow n'l', \text{Na}^+)$, where $nl=n'l'$. Wave functions for use in Eq. (1) are taken from Hartree-Fock solutions for the ground states of Ne^0 as given by Worsley¹⁷ and for the ground states of Na^{1+} as given by Hartree and Hartree.¹⁸ On subtracting from unity the transition probability for an electron to remain in its initial state, one gets the probability for transitions in which the electron may either be in some "ground state"¹⁹ other than the initial state, in some discrete excited state, or in the continuum. The values of $(1-P_i)$, where P_i is defined as $P(nl, \text{Ne}^0 \rightarrow nl, \text{Na}^+)$, are given in the third column of Table III. Transition probabilities to ground states other than the initial state are calculated from the squares of the overlap

⁹ For example, the charge spectrum resulting from the decay of Ar^{37} as taken with the field-free source volume (reference 6) compares quite well with that taken with the ion-collection source volume: A. H. Snell and F. Pleasonton, *J. Phys. Chem.* **62**, 1377 (1958).

¹⁰ For a discussion of the effect of recoil energy on electron shake-off during β^- decay, see reference 3.

¹¹ E. L. Feinberg, *J. Phys. (U.S.S.R.)* **4**, 423 (1941).

¹² A. Migdal, *J. Phys. (U.S.S.R.)* **4**, 449 (1941).

¹³ J. S. Levinger, *Phys. Rev.* **90**, 11 (1953).

¹⁴ H. M. Schwartz, *J. Chem. Phys.* **21**, 45 (1953).

¹⁵ A. E. S. Green, *Phys. Rev.* **107**, 1646 (1957).

¹⁶ For a discussion of the sudden approximation, see for example, L. I. Schiff, *Quantum Mechanics* (McGraw-Hill Book Company, Inc., New York, 1955), 2nd ed., pp. 217-220.

¹⁷ B. J. Worsley, *Can. J. Phys.* **36**, 280 (1958).

¹⁸ D. R. Hartree and W. Hartree, *Proc. Roy. Soc. (London)* **A193**, 299 (1948).

¹⁹ In this paper the $1s$, $2s$, and $2p$ orbitals are defined as ground states.

TABLE III. Single electron transition probabilities to various studies of Na⁺ during the decay of Ne²³.

Initial state (<i>nl</i>)	Single electron transition probabilities (in %)					
	$2(2l+1)$	$(1-P_i)$	Ground state (excluding initial state)	Excited state	Continuum (P_{ni})	$2(2l+1)(P_{ni})$ (%)
1 <i>s</i>	2	0.69	0.21	0.48	0.48	0.96
2 <i>s</i>	2	2.52	0.13	2.39	2.39*	4.78
2 <i>p</i>	6	3.05	none	3.05	2.70	16.2

* Includes transitions to excited discrete states, which will subsequently undergo autoionization.

integrals between the 1*s* and 2*s* states, and are given in column four of Table III. Actually, since the 1*s* and 2*s* states are already filled, there can be no transitions to these states, unless multiple shake-off occurs. The calculated values of $(1-P_i)$, however, do not include consideration of the Pauli principle and must be corrected in order to yield the transition probabilities to the excited states. This is done by taking the difference between the values given in columns three and four of Table III. The results of these differences are the transition probabilities to excited states; they are given in column five.

We now need to concern ourselves with whether the excited states will result in ionization. We shall examine, in order, transitions to excited states from the 1*s*, 2*s*, and 2*p* states.

Essentially all transitions from the 1*s* state to excited states are to the continuum. This assumption is made since the next higher state, 2*s*, is not an excited state, and the probability for transitions to still higher discrete states drops off quickly.

Ionization will occur regardless as to whether the 2*s* electron goes to a discrete excited state or to the continuum, because a 2*s* electron going to a discrete excited state will usually undergo autoionization, as will be pointed out in the next section on Auger effects.

To estimate the probability for an electron initially in a 2*p* state to go to the continuum, we have made use of experimental data on Ne²³. The argument is as follows: The probability for removing at least one electron, that is, the abundance of all the ions except charged ones, has been found experimentally to be 20.9% (see Table IV). If we subtract from 20.9% the probabilities for ionizing at least one electron from an *s* state, as given in the last column of Table III, we have remaining 15.2% as the sum of transition probabilities for going to continuum

p states. To this value must be added the probability for cases in which both *s* and *p* electrons are removed, which is about 1.0% as determined from the calculations on multiple electron shake-off. Since the probability for going to an excited *p*-state is 18.3% (that is, six times the probability, as given in Table III, column 5, for one of the 2*p* electrons to vacate its initial state), it appears that most vacancies in the initial 2*p* state result in transitions to the continuum. This is in agreement with the calculations on Kr⁷⁹ made by Green,¹⁵ who also found that vacancies usually resulted in transitions to the continuum.

Results of the calculations for single electron shake-off following the decay of Ne²³ are given in Table III. In column one are the initial states of the electrons. The number of electrons in each state are given in column two. The values in columns three, four, and five have been previously defined. Column six gives the transition probabilities to continuum states. Note that the entry for a 2*s* electron going to the continuum also includes transitions to discrete excited states, since they subsequently undergo autoionization. The transition probabilities for a given state as listed in columns 3-6 are for only one of the electrons as distinguished from the others. A fairly good approximation of the total transition probability for a given state is obtained by multiplying the values in columns 3-6 by the number of electrons in that state. In the case of the transition probability to the continuum this has been done and the results are given in column seven.

B. Auger Effect

Ionization may take place without a direct transition to a continuum state. This is possible by having a transition to a virtual state followed by an Auger process. For such an Auger process to be significant, two requirements must be met. First, an atom must have a vacancy in one of its shells and two orbital electrons such that the energy given up by having one of the electrons fill the vacancy is greater than the binding energy of the other; and, second, the probability for an Auger process must be comparable with or greater than that for a radiative process. The latter requirement is easily met for a low *Z* element like sodium since the probability for radiative transitions as opposed to Auger processes is of the order $10^{-6}Z^4$ for *K* vacancies and

TABLE IV. Charge spectrum of Na²³ ion formed from the decay of Ne²³ (% abundance).

Charge	Experiment	Theory
1	79.1	78.1
2	17.5 ± 0.1	18.9
3	2.85 ± 0.06	2.7
4	0.48 ± 0.02	0.29
5	0.081 ± 0.007	0.021
6	0.006 ± 0.002	0.0016
7	0.0008 ± 0.0008	0.00008

$10^{-8}Z^4$ for L vacancies²⁰ where Z is the nuclear charge. The energy requirement can be tested from the binding energies for Na^{1+} given by Hartree and Hartree,²¹ and from the fact that the binding energy of the first-excited state in Na^{1+} is about 14 eV.²² From the above information one sees that a Na ion will undergo autoionization following the beta decay of Ne^{23} if: (1) A vacancy is created in the $1s$ state and there are at least two electrons in some discrete state other than the $1s$ level; (2) an electron initially in a $2s$ orbital vacates that state for some discrete excited s state and there is at least one other electron in a discrete p state; or (3) Two electrons both vacate their original states for some discrete excited states.

Auger processes contribute only a small amount to the total ionization that results from the decay of Ne^{23} . Nevertheless, consideration of these processes is necessary for calculating the charge spectrum. Fortunately, for the case of neon, it is not too difficult to add in these contributions.

C. Multiple Electron Shake-Off

To compute the charge spectrum that would be expected to result from the β^- decay of neon, let us consider the probabilities for shaking one or more electrons to either discrete or continuum states. Then let us note the subsequent Auger processes that might occur.

We begin by calculating the probabilities for multi-transitions to the continuum, which are simply calculated from the products of the probabilities for single electron shake-off. The calculation for multiple shake-off assumes that the probability for shaking off a given electron is not strongly dependent on what happens to the other electrons, and thus may be approximated by the values in Table III, column 6, regardless of how many other electrons are removed. This assumption is the most important one in the calculation of the charge spectrum, and the agreement between the calculated and experimental values is largely a test of the independence of electron shake-off.

The probability of multiple shake-off of N electrons is the summation of all possible sets where N electrons are removed. The probability of each set is approximated by the product of N factors, where each factor is made up of the probability of removing a single electron from an nl orbital to the continuum, (P_{nl}), times the number of ways in which electrons can be removed. That is,

$$P(N) = \sum_{\alpha} P_{\alpha}. \quad (5)$$

Alpha is the set of numbers, $M(nl, \alpha)$, where $M(nl, \alpha)$ is the number of electrons shaken out of the nl shell, with the restrictions that $\sum_{nl} M(nl, \alpha) = N$ and $M(nl, \alpha) \leq N_{nl}$, where N_{nl} is the number of electrons originally in the orbital. P_{α} is thus expressed as

$$P_{\alpha} = \prod_{nl} N_{nl}(N_{nl}-1) \cdots \times [N_{nl} - M(nl, \alpha) + 1] P_{nl}^{M(nl, \alpha)} \times \frac{1}{[M(nl, \alpha)]!}. \quad (6)$$

Similar expressions could be derived for multiple transitions to excited discrete states, although it is not necessary for the present calculation. To complete the calculation for multiple ionization, we include the following considerations: (1) One charge is added whenever a $1s$ vacancy can result in autoionization. (2) Transition of a $2s$ electron to a discrete excited state is always considered to result in ionization. (3) The contribution of having two electrons go to discrete excited states is negligible. (4) The probability of ionizing just N electrons and no more is equal to the probability of ionizing N electrons minus the probability of ionizing $N+1$ electrons.

The calculated values for the charge spectrum following the decay of Ne^{23} are listed in Table IV. The calculated values for the lower charged ions agree very well with the experimental data. The calculation of the charge spectrum increasingly underestimates the amount of electron shake-off as the charge of the ion increases, but, in general, it seems as though multiple shake-off can be described reasonably well with rather simple assumptions.

V. CONCLUSION

Measurements have been made of the charge spectrum of sodium ions that result from the beta decay of Ne^{23} . The results afford a good chance to examine multiple electron shake-off. The probabilities for multiple electron shake-off have been computed from the product of the probabilities for single electron shake-off. As a first approximation this method proves satisfactory. The calculated values, in general, agree well with the experimental ones, although the computations underestimate the abundance of the more highly charged ions.

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²⁰ E. H. S. Burhop, *The Auger Effect* (Cambridge University Press, Cambridge, England, 1952), pp. 49 and 54.

²¹ The binding energies for the different levels in the ground state of Na^+ are calculated by Hartree and Hartree (reference 18) as follows: $1s$, 1103 eV; $2s$, 83.3 eV; and $2p$, 48.7 eV.

²² R. F. Bacher and S. Goudsmit, *Atomic Energy States* (McGraw-Hill Book Company, Inc., New York, 1932), p. 309.