Theoretical Analysis of the Charge Distribution on the Recoil Cl Ions from K Capture in A^{37} [†]

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An analysis of the charge spectrum of the recoil Cl ions which result from K capture in A³⁷, recently measured by Kofoed-Hansen, is made using recently computed Auger transition rates. It is shown that this charge spectrum, a previously measured fluorescence yield of 0.1 in Cl, and a previously measured K/Lcapture ratio of 0.92/0.08 in A³⁷ are not consistent.

OFOED-HANSEN has recently investigated¹ the old N distribution-in-charge of the recoil chlorine ions from the nuclear reaction $A^{37} + e^{-} \rightarrow Cl^{37} + \nu$ (K or L capture). The refined experimental techniques used permitted observation of the relative abundances of the variously charged recoil ions (Column A, Table I).

Recent measurements by Snell and Pleasonton² agree with Kofoed-Hansen's results in that doubly or quadruply charge ions seem to occur about half as frequently as triply charged ions. However, a preliminary estimate of the number of single-charged ions occurring appears to be less than that found by Kofoed-Hansen.

According to Kofoed-Hansen, 65 ± 5 percent of the captures are accompanied by a K-series Auger electron. Since the K-series fluorescence yield of chlorine is about 0.1³ initial K vacancies in the recoil atoms are about 90 percent efficient for the production of such electrons. Hence, the above observation implies that a fraction $p_K \sim 0.65/0.9 \sim 0.75$ of the chlorine atoms are formed with a vacancy in the K-shell.

However, Pontecorvo, Kirkwood, and Hanna⁴ have reported that the reaction in question proceeds by Kcapture with probability ~ 0.92 . If this be so, then a fraction 0.17 of the recoil atoms with K-vacancies

TABLE I. Distribution-in-charge of the recoil chlorine ions.

	Theoretical relative abundances for various distributions of initial vacancies in the recoil atom ^a			
Charge on final ion	A Experimental relative abundance ^b	$B \\ 0.75K \\ 0.25L_1$	С 0.75К 0.08L1 0.17L11,111	D 0.92K 0.08L1
1	0.26 ± 0.03	0.10	0.27	0.11
2	$0.13 {\pm} 0.04$	0.31	0.13	0.16
3	0.38 ± 0.04	0.39	0.39	0.49
4	0.18 ± 0.02	0.15	0.15	0.18
5	0.04 ± 0.01	0.04	0.04	0.05
6	0.01 ± 0.01	0.00	0.00	0.00
Average charge	$2.64 {\pm} 0.08$	2.69	2.50	2.87

^a See reference 9. ^b See reference 1.

Assisted in part by the Office of Naval Research.

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¹O. Kofoed-Hansen, Phys. Rev. 96, 1045 (1954).

⁴ Pontecorvo, Kirkwood, and Hanna, Phys. Rev. 75, 982 (1949).

 $(0.92-0.75\sim0.17)$ are unaccounted for in the Kofoed-Hansen experiment.

A possible process that might account for the missing K-vacancies by transferring them into L-vacancies is "shake-up". If the K capture occurs in a time short compared to atomic periods, the electronic wave function (which was an eigenfunction appropriate to the original nuclear charge) cannot change appreciably and will have the same shape after the capture. However, it will not be an eigenfunction appropriate to the new nuclear charge but will be instead some linear combination of such eigenfunctions. One of these eigenfunctions represents the residual atom with an L-vacancy rather than a K-vacancy. Order of magnitude estimates of the probability of this effect render it highly suspect as an explanation of the missing K-vacancies. In the first place, present theoretical estimates of such a process^{5,6} are too small to account for such a large fraction of the events. In the second place, even if such shake-up did occur, the electron captures in question would have been indistinguishable from L captures in the Pontecorvo, Kirkwood, and Hanna (PKH) experiment, and would have been reported as such, since the energy given up by the atom during shake-up is carried off by the neutrino and the recoil ion which thus have energies appropriate to L rather than K capture.

The above discrepancy between the PKH measurements and the Kofoed-Hansen experiment further manifests itself in the latter's distribution-in-charge. The appearance of multiply charged chlorine ions in that experiment is due to the reorganization of the atom with an original K- or L-vacancy formed in the nuclear reaction by subsequent Auger effects,^{7,8} each Auger process contributing one unit of positive charge to the final ion.

A new theoretical treatment of the Auger effect⁹ using the techniques available with modern high-speed digital computers has been carried out for argon (Z=18), krypton (Z=36), and silver (Z=47). A Hartree selfconsistent field method was used to obtain better approximations to the electronic wave functions in atoms

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 Broyles, Thomas, and Haynes, Phys. Rev. 89, 715 (1953).

⁵ J. S. Levinger, Phys. Rev. 90, 11 (1953).
⁶ A. Migdal, J. Phys. (U.S.S.R.) 4, 423 (1914).
⁷ M. L. Perlman and J. A. Miskel, Phys. Rev. 91, 899 (1953).
⁸ E. P. Cooper, Phys, Rev. 61, 1 (1942).
⁹ R. A. Rubenstein and J. N. Snyder (to be published).

TABLE II. Transition probabilities in argon calculated from theory.^a

$P_{k}(K \rightarrow I_{k}, m) = 0.122$	$P(K \rightarrow M) = 0.000$			
$P(K \rightarrow L_1^2) = 0.061$	$P(K \rightarrow L_T M) = 0.009$			
$P(\overline{K} \rightarrow L_{\rm I} L_{\rm II, III}) = 0.199$	$P(K \rightarrow L_{\text{II, III}}M) = 0.082$			
$P(K \rightarrow L^2_{\text{II, III}}) = 0.498$	$P(L_{\rm I}, \rightarrow L_{\rm II}, \Pi M) = 1.00$			
$P(L_{\rm II, III} \rightarrow M^2) = 1.00$				

Probabilities marked with the subscript r represent radiative transitions. All others are Auger processes. K stands for a vacancy in the K shell, $L_{\rm I}$ for a vacancy in the $L_{\rm I}$ shell, $L_{\rm II, III}$ for a vacancy in either the $L_{\rm II}$ or $L_{\rm III}$ shell, and M for a vacancy in any M shell.

^a See reference 9.

with vacancies. With these wave functions the matrix elements for Auger transitions and radiative transitions were computed.

The results of these computations have been expressed in Table II as a list of probabilities for the several transitions. Each entry is the ratio of the transition rate for the given process to the total rate for *all* processes originating in the same level. Only the important transitions are shown in Table II.

The resulting set of Auger and radiative transition probabilities makes it possible to compute the relative probability of occurrence of the different charged ions that can arise as a result of Auger cascades starting from either an initial K-vacancy or an initial L-vacancy. Column D of Table I gives the charge distribution to be expected if the initial vacancy distribution is 0.92K, $0.08L_{\rm I}$ as measured by PKH. This is inconsistent with Kofoed-Hansen's observed charge distribution.

However, it can be argued from the Kofoed-Hansen data and from reasonable assumptions about the atomic reorganization process, that the above discrepancy cannot be explained simply by a reassignment of the L/K ratio such as to yield K capture with 75 percent probability, L capture with 25 percent probability, i.e., that the unexplained 17 percent of the recoil ions could not have been formed in a state with an $L_{\rm I}$ -vacancy as would necessarily have been the case were they products of L-capture reactions (the subscript indicates in which subshell of the L shell the vacancy occurs). The recent theoretical investigation⁹ of the Auger effect shows that it is reasonable to assume:

(1) Almost every $L_{\rm I}$ -vacancy is filled by a Coster-Kronig transition¹⁰ of the type $L_{\rm I} \rightarrow L_{\rm II, III} M$, in which an $L_{\rm I}$ -vacancy is destroyed while an *M*-vacancy and either an $L_{\rm II}$ - or an $L_{\rm III}$ -vacancy is created with the ejection of an Auger electron. The matrix element for this transition is anomalously large compared with ordinary Auger transitions because of considerable overlap between the 2s and 2p electronic functions.^{9,10} This transition is energetically allowed in chlorine,⁸ and the assumption that it occurs with unit probability is borne out by the excellent agreement of the theoretical with the experimental abundance for triply and quadruply charged ions (Table I, columns A, B, and C). If this assumption were incorrect, the theory would lead to underestimate of these abundances.

(2) Every $L_{II,III}$ -vacancy is filled by an Auger process, since radiation rates are much smaller than Auger transition rates in the L shell.

(3) No mechanism exists whereby an M-vacancy may be filled by an Auger process.¹¹

The above arguments are relatively insensitive to the numerical values of the theoretically computed Auger transition rates; if they be granted, then a chlorine atom which is formed with an L_{I-} or $L_{II, III}$ -vacancy will always lead to a doubly- (or singly-) charged final ion.

Thus, if the K and L captures occur with relative probability 0.75 and 0.25, respectively, then one would expect to observe doubly-charged ions with at least 0.25 probability since L capture involves only the $L_{\rm I}$ electron. (The theoretical distribution based on an L/Kratio of 0.25/0.75 is shown in Table I, column B.) Kofoed-Hansen observes doubly-charged ions only 13 percent of the time.

The Kofoed-Hansen experiment agrees well with the theory if one assumes that all of the extra 17 percent of the recoil atoms lead to singly-charged final ions (e.g., if they are formed with an $L_{II, III}$ -vacancy). This case is shown in Table I, column C.

It can therefore be concluded that the Kofoed-Hansen experiment seems to contradict the earlier PKH measurement of the L/K ratio; on the other hand, a simple revision of this ratio is not in itself sufficient to explain the observed distribution-in-charge of the final ions.

¹⁰ E. H. S. Burhop, *The Auger Effect* (Cambridge University Press, Cambridge, England, 1952).

¹¹ The possibility that surface effects in Kofoed-Hansen's apparatus may have degraded some of the higher charged ions into lower charged ones has been neglected.