PHYSICAL REVIEW C VOLUME 25, NUMBER 5 MAY 1982

Electron capture shakeoff

Akira Suzuki and J. Law
Guelph Waterloo Program for Graduate Work in Physics, Guelph Campus, University of Guelph, Guelph, Ontario, Canada, N1G 2W1
(Received 22 January 1982)

K-electron shakeoff probabilities during *K* capture have been calculated using Dirac-Fock-Slater wave functions, and by direct calculation of the ejected-electron continuum wave functions. The results indicate an overall better agreement with experimental results, but leave room for some residual electron-electron correlation effects.

[RADIOACTIVITY *K*-vacancy creation calculations in electron capture.]

I. INTRODUCTION

There have been many theoretical and experimental investigations of the atomic internal ionization phenomena caused by the sudden change of nuclear charge during various nuclear decay processes since the pioneering works by Feinberg[1,2] and Migdal[3] in 1941. Most of the history can be traced in the review articles by Freedmann,[4] and Walen *et al.*[5]

In the ionization process, an electron in a bound state of the parent atom can be ejected into a continuum state (shakeoff) or promoted into higher excited states (shakeup) of the daughter atom, leaving a vacancy in the corresponding orbit.

The present paper is concerned with the double *K*-vacancy creation process by *K*-electron shakeoff when *K*-electron capture occurs. The evaluation of the double *K*-vacancy creation probability involves the calculation of the overlap integral between the initial state corresponding to the parent atom and the final state which has an ejected electron in a continuum state and double vacancies in the *K* shell of the daughter atom. Most of the theoretical efforts have concentrated on the treatment of the wave functions contained in the overlap integral, because of the significant sensitivity of the result to the choice of wave functions.

Following Feinberg and Migdal, there have been two essentially different approaches for the evaluation of the overlap integral, i.e., the Primakoff-Porter method[6–9] and the Coulomb propagator method.[10–12] Both models take as their starting points the use of hydrogenic wave functions and include an attempt to take account of the initial-state correlation between the two *K* electrons. Whether the seeming satisfactory agreement of the numerical results with the latest experimental data will be altered when more precise wave functions are used, as well as inclusion of final state correlations, is not known. In an attempt to answer these questions, we have applied our self-consistent-field (SCF) method to this problem. The latter has also been successfully applied to the calculation of the *K*-electron shakeoff probability in β decay and gives good agreement with the experimental data.[13] (Hereafter this article is referred to as I.)

In Sec. II, we briefly present the important differences between the above approaches. In Sec. III, we introduce the SCF method for the overlap integral with a few numerical calculations for comparison with previous methods. All numerical results of probability are given in Sec. IV together with discussions concerning the discrepancies between our results and experimental data.

II. TREATMENT OF OVERLAP INTEGRAL

The formulation of the double *K*-vacancy creation probability by electron capture (EC) has been given in several articles in detail. Here we follow the notations used in the article by Law and Campbell (LC) (Ref. 11) and give only final expressions to help with the discussions. We consider only the shakeoff process in the double *K*-vacancy creation process. We expect from the correspond-

ing results in I that the shakeup contribution to the probability will be considerably smaller, so as to be negligible in comparison with the shakeoff process.

The theory for K shakeoff in K-electron capture is based upon the sudden approximation.[1-3] Owing to the change of nuclear charge by the orbital (K-shell) electron capture, the whole atomic system undertakes a rapid transition from the ground state of the parent atom to the final state, emitting the other K electron to the continuum and leaving the double-K-vacancy state in the daughter atom. The corresponding decay rate is derived following the Fermi "golden rule" as

$$\lambda(S0)=\frac{4}{\pi}G_v^2\xi\,|\,\phi_K(0)\,|^2\int\frac{d^3p}{(2\pi)^3}\,|\,\langle e_p'\,|\,e_K\rangle\,|^2\times[W_0-B_K(f)-W_p]^2\,, \tag{1}$$

assuming the usual EC Hamiltonian and the Slater determinants for the initial and final states. The various exchange processes involving higher orbits may also be considered fully by evaluating the Slater determinants. According to our estimates, the contribution from them is less than 1% of the shakeoff probability so that they may be neglected. The normal EC decay rate, in which the electron in the K shell of the parent atom remains in the daughter-atom K shell to form the final state with one K vacancy, is

$$\lambda(\mathrm{EC})=\frac{4}{\pi}G_v^2\xi\,|\,\phi_K(0)\,|^2(W_0-1)^2\,. \tag{2}$$

The measurements of the double-K-vacancy creation probability yield the probability per capture, i.e.,

$$P_{KK}(S0)=\lambda(S0)/\lambda(\mathrm{EC})\,. \tag{3}$$

One of the difficulties of the problem lies in the evaluation of the overlap integral in Eq. (1). The earliest approach was given by Primakoff and Porter (PP) in 1953.[6] They used nonrelativistic hydrogenic wave functions in the overlap integral, in which the mutual correlation effect between two K-shell electrons and the screening effect in the initial state are taken into account independently in terms of the phenomenological effective-charge parameters. These parameters were fixed to reproduce the Hylleraas variational wave function for a two-electron atom. The extension of the PP theory into the relativistic scheme was made by Stephas,[7] Mord,[8] Law, and Campbell,[11] and Mukoyama *et al.*[9] In the Mukoyama *et al.* paper, the Dirac wave functions were shielded according to effective charges extracted from the SCF calculation by Carlson *et al.*[14] Although these authors[9] gave numerical results for atoms for which experimental data existed, as pointed out by Intemann,[15] the probabilities are too sensitive to pertinent parameters to draw a meaningful quantitative conclusion. However, this sensitivity indicated the importance of initial correlations and called for the theory to remove the ambiguity with respect to phenomenological parameters.

In the alternative approach developed by Intemann and Pollock (IP),[10] and also by Law and Campbell (LC),[11] the initial state is determined perturbatively to introduce the K-shell correlation in terms of the Coulomb propagator function. Therefore, as far as the initial K-electron correlation is concerned, this approach is essentially parameter free. The apparent large discrepancy existing in the numerical results of IP and LC was ascribed to the truncation of the perturbation expansion with less terms in LC treatment, as pointed out by Intemann.[12]

So far, most attention in evaluating the overlap integral has been paid to the initial-state correlation, and the final state has been considered as the scattering state by the nuclear charge of the daughter atom. It has been expected that the screening effect on the continuum electron in the final state may be compensated for by the effect of two K vacancies. This is only qualitatively true and may not be so quantitatively.

In paper I, we discussed the shakeoff process in β decay. In our approach, the initial state was produced by solving the Dirac-Fock-Slater (DFS) SCF equation. The final continuum state was determined numerically as the scattering state defined by the SCF potential describing the final state. The latter depends strongly on the final atomic configuration. The results showed that the problem is complicated by effects coming from the many-body nature, so that it cannot reliably be reduced to a simple parametrization by choosing initial and final shielding charges. The SCF calculation gave good agreement with experimental data. From the results on β^+ decay, which is the alternate process to EC decay, we observe that: (i) The screening in the initial state, which contains the major part of correlation between the two K-shell electrons as determined through the SCF procedure, works to reduce the probability by pushing the K-shell wave function out to give rise to less overlap with the final

state. (ii) The screening in the final state, which acts on continuum electrons as an effective repulsive potential, tends to increase the probability. (iii) The vacancies in the K shell of the final state have a tendency to reduce the probability, since they act on the continuum electron as an additional effective attractive potential. The effects (i) and (ii) appear to have nearly the same magnitude, but both are much stronger than the effect (iii). Therefore, if these features are applied to EC decay, we should see similar trends in older numerical calculations. Thus, when we apply hydrogenic wave functions to the initial and final states, e.g., as in the Dirac-wave function calculation, we will overestimate the probability. When we introduce the initial-state correlation into the initial hydrogenic wave function but leave the final state uncorrelated and unshielded, e.g., as in the PP or the Coulomb propagator methods, we will underestimate the probability. In order to show that we observe these effects, the numerical results of previous calculations are shown in Table I. We have taken the results of Mukoyama *et al.*[9] as the equivalent (relativisitic) PP method, and those by Intemann[10] as the Coulomb propagator method. The values corresponding to the Dirac wave function calculation without correlations were quoted from the paper by LC.[11] This trend suggests that it may be valid to apply our SCF approach from beta-decay shakeoff to this problem.

TABLE I. Previous calculations of the shakeoff probability (in multiples of 10^5). The results were given by Law and Campbell (LC) for the Dirac wave function, by Mukoyama *et al.* (MIKS) for the relativistic PP method, and by Intemann for the Coulomb propagator method.

	Exp.	LC	MIKS	Intemann
Ar	44 ±8	115.50	14.2	21.12
	37 ±9			
Fe	38 ±7	48.36	8.81	8.26
	28			
	12 ±4			
	10.1 ±2.7			
Ge	13.3 ±1.4	29.83	4.56	4.72
	13 ±5			
	24			
Cs	5.0 ±1.0	8.69	0.709	0.92
	2.5 ±0.2			
	2.0 ±1.3			
	1.33±0.33			
	2.3 ±0.3			
	1.4 ±0.1			
Er	1.5 ±0.4	4.77	0.304	0.39
	0.67±0.39			

III. ELECTRONS IN BOUND AND CONTINUUM STATES

As shown in Sec. II, the introduction of the initial-state correlation into the Dirac wave function reduces the shakeoff probability by 80–90%. On the other hand, if the final-state correlation is appreciable as a consequence of compensation between the two effects of screening and the K vacancies, this will reduce the contribution to some extent. The result depends on this delicate cancellation.

In principle, these effects are indistinguishable, coming from the many-body character of the system. Therefore we should treat the problem in a self-consistent way. Under the SCF scheme, the initial state is regarded as the bound state produced by the SCF one-body central potential which emerges from the DFS theory. The continuum electron state is assumed to be the scattering state produced in a similar SCF potential of the daughter atom with the proper atomic configuration after EC decay. The correct choice of the atomic configuration in creating the SCF potential is crucial for this kind of problem as seen in paper I. In order to stress this, we give in Table II values of P_{KK} for Ge EC decay. These values were calculated with the SCF potentials that resulted from the use of three different atomic configurations in the final state: (a) two K vacancies, (b) one K vacancy, and (c) the ground state for the daughter atom Ga, with the configurations

(a) $\{[\mathrm{AR}]1s^{-2}3d^{10}4s^{2}4p^{1}\}$,

(b) $\{[\mathrm{AR}]1s^{-1}3d^{10}4s^{2}4p^{1}\}$,

(c) $\{[\mathrm{AR}]3d^{10}4s^{2}\}$.

TABLE II. The shakeoff probabilities (in multiples of 10^5) for Ge decay calculated with the SCF potentials emerging from (a) two K-shell vacancies, (b) one K-shell vacancy, and (c) ground-state atomic configurations of Ga. For comparison the result with unshielded Dirac wave functions is given in column (d).

	Exp.	(a)	(b)	(c)	(d)
Ge	13.3±1.4	11.12	21.96	42.67	6.94
	13 ±5				
	24				

We note that the calculation with respect to (b) corresponds to the application of Carlson *et al.*'s theory in beta decay to EC decay with neglect of the small contribution from the shakeup process. It is clear that the choices of the atomic configurations (b) and (c) result in unreasonably large shakeoff probability compared with the measured result. This trend appears common to all elements. Furthermore, for the purpose of confirmation that the screening effect in the final state will not be compensated by the effect from the vacancies, we also performed a calculation using Dirac wave functions for the final state but with the initial state obtained from the DFS theory. This result is given in column (d) of Table II. As seen clearly from comparison between (a) and (d), the simultaneous neglect of screening and the vacancies leads to reduction of the probability nearly by a half. We can ascribe the main reason of underestimation, which commonly appeared in the numerical results in past works, to lack of the the final-state correlation. The numerical results for elements for which experimental data are available at present are given in Sec. IV. There, the SCF potentials with the proper choice of atomic configurations in the final state, i.e., those with two K vacancies in the daughter atom, are used to describe the scattering wave function for the continuum electron.

IV. NUMERICAL RESULTS AND DISCUSSION

As in paper I, we used the program of Lindgren and Rosén[16] to produce the initial K-shell bound-state wave function and the SCF potential for the scattering wave function calculation in the final state. The numerical results for those elements for which experimental data are available at present are listed in Table III. This table has three columns, depending on the approximation used for the ex-

TABLE III. Shakeoff probabilities (in multiples of 10^5). See text for description of the various columns. $B_K(i)$ and $B_K(f)$ represent the K-shell binding energies of the parent and the daughter atoms, respectively.

	E (keV)	$B_K(i)$ $B_K(f)$ (keV)	P (exp)	P (ODFS)	P (LDA)	P (AKF)	Ref.
Ar	813.8	3.21	44 ± 8	49.36	52.94	55.40	17
		2.82	37 ± 9				18
Fe	231.4	7.11	38 ±17	18.80	20.06	21.14	19
		6.54	28				20
			12 ± 4				21
			10.1 ± 2.7				22
Ge	235.7	11.10	13.3 ± 1.4	11.12	11.84	12.55	23
		10.37	13 ± 5				24
			24				25
Pd	507.2	24.35	3.13± 0.31	5.59	6.03	6.45	26
		23.22					
Cd	94.0	26.71	72^{+5}_{-15}	0.83	0.89	0.97	27
		25.51	15.2 ± 2.4				28
			2.8 ± 0.7				29
			1.02± 0.36				30
Cs	355.6	35.99	5.0 ± 1.0	2.99	3.22	3.48	31
		34.57	2.5 ± 0.2				32
			2.0 ± 1.3				33
			1.33± 0.33				34
			2.3 ± 0.3				35
			1.4 ± 0.1				38
Er	377.1	57.49	1.5 ± 0.4	1.58	1.71	1.88	36
		55.62	0.67± 0.39				34
Bi	772.4	90.53	0.6 ± 0.25	1.81	1.97	2.20	37
		88.01					

change interaction part in the SCF potential so as to create a more realistic representative potential for particles in the continuum state, namely the use of the unmodified potential (ODFS) [column labeled P (ODFS)], the use of the modified SCF potential using the local density approximation (LDA) [column labeled P (LDA)], and the use of the modified SCF potential by the average Fermi momentum approximation (AKF) [column labeled P (AKF)]. These are the same approximations as used in I for the choice of potential for the description of the continuum electron.

All calculations for the forbidden decays were done without the shape factors, since we expect that the introduction of the shape factor will not change the probability significantly as seen in the case of beta-decay shakeoff. From Table III, it can be seen that the theoretical calculations show reasonable agreement with the experimental data except for the cases of Pd and Bi and the recently measured case of Cs.

The results for Pd and Bi decays, however, may suggest a breakdown of the model. The large theoretical values obtained for Pd and Bi mean that the electron in the final continuum state will feel a more attractive potential and/or that the electron in the initial bound state will feel a more repulsive potential than obtained from the SCF theory. From the fact that most of the theoretical calculations including those of the β-decay shakeoff show good agreement with the experimental data, it appears that the SCF potential can simulate to a considerable extent the "true potential" which can describe the "exact electron state" in the initial and final states for these systems. Therefore we may expect that the deviation of the SCF potential from the true potential is small, probably less than the variation of the SCF potential due to an increase or decrease by a unit of nuclear charge, as evidenced by the successful application of the model to the β-decay case. Thus, if the theory is valid, we may ascribe the source of the extra repulsion in the initial state and/or the extra attraction in the final state to the choice of the atomic configuration or to the SCF procedure.

Concerning the final state, in order to produce the SCF potential we adopted the atomic configuration with two K vacancies. This is the most favored one for maximum attraction.

On the other hand, in the initial state, it may be expected that the residual correlation between two K-shell electrons on the one-particle basis of the Dirac-Fock theory may be a source of extra repulsion. This can be simulated in terms of the correlated K-shell wave function

$$\Phi_K(\vec{r}_1,\vec{r}_2)=A\{\phi_\uparrow(r_1)\phi_\downarrow(r_2)f(|\vec{r}_1-\vec{r}_2|)\}\ , \tag{4}$$

where A is an antisymmetrization operator including an appropriate normalization. The correlation function is chosen as

$$f(r)=\exp(\gamma r/2)\ , \tag{5}$$

where γ may be interpreted as the effective charge coming from the residual repulsive correlation between two K-shell electrons, which may be expected to be considerably smaller than unity. In Fig. 1 we show the γ dependence of the shakeoff probability for Bi decay as an example. The values of γ which can reproduce the measured probability within the measurement error lie between 0.6 and 1.0. We feel, from the above discussion, that this range is somewhat larger than expected.

If the introduction of the residual initial-state correlation is necessary to describe the situation more precisely, then similar considerations must apply to all cases including β decay, although the size

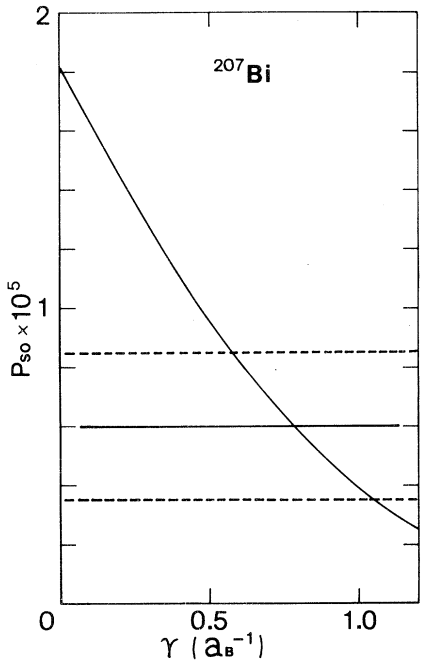

FIG. 1. The γ dependence of the shakeoff probability in Bi decay. The parameter γ is given in inverse atomic units of length (Bohr^{-1}). The corresponding experimental datum is $(0.6\pm0.25)\times10^5$, which is shown by the horizontal solid line and two dotted lines representing the range of the experimental errors.

of γ may be different in each case. We have to note, however, that the discrepancy existing between the theoretical calculation and the experimental data for Cu β^+ decay, which is the only available measurement of K-shell electron shakeoff in β^+ decay, suggests an opposite trend, namely we need more attraction in the initial state and/or more repulsion in the final state. Therefore the residual initial-state correlation makes the situation of Cu β^+ decay worse, unless we have a negative effective charge meaning an attractive correlation only for this case.

We hope that reconfirmation of the K-shell shakeoff measurements for Pd and Bi will be forthcoming as they may provide a direct indication of residual correlations in these atoms. The most recent measurments on Cs seem to support this conjecture.[38]

ACKNOWLEDGMENTS

We would like to thank the Natural Science and Engineering Research Council of Canada for a grant in aid of this research, and the University of Guelph for providing the computing facilities and computer time on the Amdhal 370/V5. Discussions with Dr. J. L. Campbell have been invaluable during the course of this work. Finally, we would like to thank Dr. I. Lindgren for the use of his Dirac-Fock-Slater program.

[1]E. L. Feinberg, J. Phys. (USSR) 4, 423 (1941).
[2]E. L. Feinberg, Yad. Fiz. 1, 612 (1965) [Sov. J. Nucl. Phys. 1, 438 (1965)].
[3]A. Migdal, J. Phys. (USSR) 4, 449 (1941).
[4]M. S. Freedmann, Annu. Rev. Nucl. Sci. 24, 209 (1974).
[5]R. J. Walen and C. Briançon, *Atomic Inner-Shell Processes I,* edited by B. Crasemann (Academic, New York, 1975), p. 233.
[6]H. Primakoff and F. T. Porter, Phys. Rev. 89, 930 (1953).
[7]P. Stephas and B. Crasemann, Phys. Rev. 164, 1509 (1967); P. Stephas, *ibid.* 186, 1013 (1969); P. Stephas and B. Crasemann, Phys. Rev. C 3, 2495 (1971).
[8]A. J. Mord, Nucl. Phys. A192, 305 (1972); A. J. Mord, Proceedings of the International Conference on Inner-Shell Ionization Phenomena, Atlanta. 1972, edited by R. W. Fink *et al.*, Georgia Institute of Technology/Emory University Report No. USAEC-CONF-7202404, 1973, p. 2079.
[9]T. Mukoyama, Y. Isozumi, T. Kitahara, and S. Shimizu, Phys. Rev. C 8, 1308 (1973).
[10]R. L. Intemann and F. Pollock, Phys. Rev. 157, 41 (1967); R. L. Intemann, *ibid.* 178, 1543 (1969); 188, 1963 (1969).
[11]J. Law and J. L. Campbell, Nucl. Phys. A199, 481 (1973).
[12]R. L. Intemann, Nucl. Phys. A219, 20 (1974).
[13]J. Law and A. Suzuki, Phys. Rev. C 25, 514 (1982).
[14]T. A. Carlson, C. C. Lu, T. C. Tucker, C. W. Nestor, and F. B. Malik, Oak Ridge National Laboratory Report No. ORNL-4614, 1970 (unpublished); Phys. Rev. 169, 27 (1968).
[15]R. L. Intemann, in *Proceedings of the International Conference on Inner Shell Ionization Phenomena—Invited Papers, 1976, Freiburg, West Germany,* edited by W. Mehlhorn and R. Brenn (Universität Freiburg, Freiburg, Germany, 1976), p. 297.
[16]I. Lindgren and A. Rosén, Case Stud. At. Phys. 4, 93 (1974).
[17]J. A. Miskel and M. L. Perlman, Phys. Rev. 94, 1683 (1954).
[18]R. W. Kiser and W. H. Johnston, J. Am. Chem. Soc. 81, 1810 (1959).
[19]G. Charpak, J. Phys. Rad. 16, 567 (1955).
[20]H. Primakoff and F. T. Porter, Phys. Rev. 89, 930 (1953). This paper not only gave the first correlation treatment, but also gave one of the earliest measurements.
[21]J. P. Briand, P. Chevallier, A. Johnson, J. P. Rozet, M. Tavernier, and A. Touati, Phys. Lett. 49A, 51 (1974).
[22]T. Kitahara and S. Shimizu, Phys. Rev. C 11, 920 (1975).
[23]M. Langevin, J. Phys. Rad. 19, 341 (1958).
[24]W. von Oertzen, Z. Phys. 182, 130 (1964).
[25]J. P. Briand, P. Chevallier, M. Tavernier, and J. P. Rozet, Phys. Rev. Lett. 27, 777 (1971).
[26]C. W. E. van Eijk, J. Wijnhorst, and M. A. Popelier, Phys. Rev. A 20, 1749 (1979).
[27]K. Knauf and H. Sommer, Z. Phys. 183, 10 (1965).
[28]H. J. Nagy, G. Schupp, and R. R. Hurst, Phys. Rev. C 11, 205 (1975).
[29]C. W. E. van Eijk and J. Wijnhorst, Phys. Rev. C 15, 1068 (1977).
[30]C. W. E. van Eijk, J. Wijnhorst, and M. A. Popelier, Phys. Rev. C 19, 1047 (1979).
[31]H. Daniel, G. Schupp, and E. N. Jensen, Phys. Rev. 117, 823 (1960).
[32]N. L. Lark and M. L. Perlman, Phys. Rev. 120, 536 (1960).
[33]K. M. Smith, Ph. D. thesis, University of Glasgow, 1964 (unpublished).
[34]H. J. Nagy, G. Schupp, and R. R. Hurst, Phys. Rev. C 6, 607 (1972).
[35]R. J. Walen, CNRS report 1976/77 (unpublished).
[36]H. Ryde, L. Persson, and K. Oelsner-Ryde, Nucl. Phys. 47, 614 (1963).
[37]J. P. Briand, J. P. Rozet, P. Chevallier, A. Chetioui, M. Tavernier, and A. Touati, J. Phys. B 13, 4751 (1980).
[38]Y. Isozumi, Ch. Briançon, and R. J. Walen, Phys. Rev. C (to be published); and private communication.