Double K-shell ionization in electron capture decay

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Using a semirelativistic theory previously developed by the author, we have computed the total probability per K-capture event for the ionization of the remaining K electron for a dozen nuclides of interest. Based on hydrogenic wave functions and accurate to relative order $(Z\alpha)^2$, the theory takes into account the correlation between the two initial K electrons and permits adjustments for screening. Numerical results exhibiting the effects of screening are presented. A comprehensive comparison of the predictions of this theory, as well as those of other theoretical models, with recent experimental data, is also given.

Inner-shell ionization during electron capture (EC) is a higher-order decay process that has long been a subject of study, both theoretical and experimental.[1] Of particular interest has been the ionization of the remaining K-shell electron during an allowed K-capture transition, a process for which P_K, the total ionization probability per K-capture event, is independent of all nuclear matrix elements but sensitive to screening and to correlation effects between the two K electrons in the initial state.

During the last thirty years a number of calculations of P_K have been reported. But prior to the early 1970's, available experimental data was very limited and often lacking in precision. Thus, there was little incentive to refine the available theories. However, since then a growing number of high-precision experiments have appeared in the literature, and it is the purpose of this paper to provide a comprehensive comparison between all recent experimental data and the predictions of the various theoretical models currently available. Preliminary to this undertaking, we shall briefly review the models which have been proposed and report on some refinements of a particular theory previously developed by the author.

Common to all theoretical studies of K-shell ionization in EC is a simplified model in which only those leptons which participate in the rearrangement process are included in the description of the leptonic states. All other electrons are regarded as inert; it is assumed that their influence on the process, largely one of shielding, can be taken into account through a suitable choice of forms for the wave functions of the participating electrons. In this approximation, the transition matrix element (for an allowed β transition) is given by

$$M=(1-P_{12})\int d\mathbf{r}\,\phi_{\mathbf{P}}^{(z')\dagger}(\mathbf{r})\,\phi_{\nu}^{\dagger}(0)\,B\,\phi_{1,2}^{(z)}(0,\mathbf{r}) \ , \tag{1}$$

where $\phi_{1,2}^{(z)}$ and $\phi_{\mathbf{P}}^{(z')}$, labeled by the appropriate eigenvalues of the nuclear charge number operator, and ϕ_{ν} are the normalized wave functions representing the two initial K electrons, the ejected electron with momentum $\mathbf{P}$, and the neutrino, respectively. P_{12} is the exchange operator which interchanges the two initial electrons and B is a 4×4 matrix containing the operative nuclear matrix elements.

The principal theoretical challenge associated with the evaluation of (1) is the construction of a satisfactory form for the wave function of the initial two-electron state, one which takes into account the correlations resulting from the mutual Coulomb repulsion between the two K electrons. To this end there have been developed two different approaches, characterized by the method by which the initial two-electron wave function is constructed.

In the variational approach, pioneered by Primakoff and Porter,[2] the electron-electron interaction is included in the unperturbed Hamiltonian, whose eigenstates are then obtained by means of the Rayleigh-Ritz method, or, in more sophisticated calculations, a self-consistent-field method. Some years later, Intemann and Pollock[3] developed a perturbation approach in which (assuming that $Z\gg1$) the electron-electron interaction is regarded as a perturbation on the nuclear Coulomb interaction. The unperturbed wave function is then simply the product of two K-shell hydrogenic wave functions; and the perturbed wave function is readily calculated to first order in the fine structure constant α by means of conventional perturbation theory. In this approximation the transition matrix element (1) separates into what are commonly called the shakeoff (SO) and direct-collision (DC) contributions:

$$M=M_{\mathrm{SO}}+M_{\mathrm{DC}} \ , \tag{2a}$$

with

$$M_{\mathrm{SO}}=(1-P_{12})\,\phi_{\nu}^{\dagger}(0)\,B\,\phi_{1}^{(z)}(0)\int d\mathbf{r}\,\phi_{\mathbf{P}}^{(z')\dagger}(\mathbf{r})\,\phi_{2}^{(z)}(\mathbf{r}) \ , \tag{2b}$$

$$M_{\mathrm{DC}}=\alpha(1-P_{12})\,\phi_{\nu}^{\dagger}(0)\,B\int d\mathbf{r}\int d\mathbf{r}'\,G_E(0,\mathbf{r})\,\phi_{1}^{(z)}(\mathbf{r})\times\frac{1}{|\mathbf{r}-\mathbf{r}'|}\,\phi_{\mathbf{P}}^{(z')\dagger}(\mathbf{r}')\,\phi_{2}^{(z)}(\mathbf{r}') \ , \tag{2c}$$

and, appearing in (2c), the Dirac-Coulomb Green's function $G_E(\mathbf{r},\mathbf{r}')$ with $E=E_1+E_2-W$, where E_1, E_2, and W are the total energies of the two initial electrons and the final electron, respectively. From this result it is clear that the amplitude M_{DC} arises from the initial-state interaction between the two K-electrons when that interaction is regarded as a perturbation on the nuclear Coulomb interaction. Indeed, because of the near orthogonality of $\phi_{\mathbf{P}}^{(z')}$ and $\phi_{2}^{(z)}$ (since $z'=z-1$), M_{SO} is also of order α as is readily demonstrated. Thus, it is logically necessary to consider both M_{SO} and M_{DC} in a lowest order calculation of M.

In all fully relativistic calculations based on the variational approach which have thus far been reported, the trial wave function has been chosen to be a direct product of single particle functions $\psi_n(\mathbf{r})$. In this case, the expression for M

given by (1) reduces to

$$M \approx M'_{SO} = (1-P_{12})\phi_\nu^\dagger(0) B \psi_1^{(z)}(0) \int d\mathbf{r} \psi_P^{(z')}(\mathbf{r}) \psi_2^{(z)}(\mathbf{r}) \ , \tag{3}$$

and has the same form as M_{SO}.[4] Thus, from the point of view of the perturbation approach, the use of variational wave functions in (3), rather than the unperturbed wave functions which appear in (2a), is an attempt to simulate in a simple way the contribution to M from M_{DC}. To judge the success of this procedure, we consider the results of several theoretical studies.

The most refined results which have thus far been obtained with the variational approach are the self-consistent-field (SCF) results of Suzuki and Law (SL).[5] In the evaluation of (3), these authors used Dirac-Fock-Slater wave functions and, to generate the scattering wave function of the ejected electron, they assumed the SCF potential corresponding to a fully relaxed daughter atom with two K vacancies. Thus, to the extent possible within the framework of the SCF approximation, these results take into account not only the initial-state interaction between the two K electrons, but also the influence of the other orbital electrons. Numerical results from this work are displayed in Table I.

With the perturbation approach a much more detailed analysis of the initial-state interaction becomes possible, but at the price of much more complex calculations. Consequently, fully relativistic results have yet to be obtained. The most refined calculation currently available is that reported some years ago by the author.[6] In this work the semirelativistic eigensolutions of the symmetric Hamiltonian of Biedenharn and Swamy,[7] a Hamiltonian which differs from the Dirac-Coulomb Hamiltonian by terms of order $(z\alpha)^2$, was employed; and numerical results for P_K were reported for a few specific nuclides.[8] Since then, there has been a significant increase in the number of high-precision experiments which have been reported, in many cases the data being found to disagree with the SL predictions by factors of 1.5–2. As a result, there has been renewed interest in the predictions of this semirelativistic perturbation (SRP) theory calculation.

This has led the author to reexamine this work. In particular, we have made some minor revisions in our computer program, and have employed more recent values for $W_0 = E - B_K(i)$, the energy released in a K-capture transition. More significantly, we have also introduced some refinements into the formalism. Specifically, we now distinguish between z and $z' = z - 1$, the charge numbers of the parent and daughter nuclides, in describing the initial and final electronic states. (This was not done in previously reported SRP calculations where, in the spirit of perturbation theory, the approximation $z' = z$ was used.) In this way the most important effect of screening on the continuum electron is taken into account.

We have also investigated the effects of screening on the two initial electrons by replacing z by an effective nuclear charge number $z_{\text{eff}} = z - \sigma$.[9] This does not violate the spirit of our perturbation approach, and compensates for the fact that by treating the electron-electron interaction as a perturbation, the theory fails to give adequate recognition to the mutual screening by the two initial K electrons. Since no account has yet been taken of the presence of $L,M, \ldots$ shell electrons, it is appropriate to include their contributions to σ as well. To do so in a reasonably realistic way, we have used the values of Froese Fischer,[10] obtained from Hartree-Fock calculations. The results obtained also are displayed in Table I, where they may be compared with the SL results. It is evident that screening effects increase the SRP predictions, particularly for the lighter nuclides, the influence of screening decreasing with increasing z as expected. However, the resulting theoretical values are still substantially smaller than the corresponding predictions of the SL theory.

It should be noted that the SRP theory, as presently formulated, is only applicable to transitions for which the decay energy is below the threshold for competing positron emission; hence, P_K could not be computed for ^{65}Zn or the 10% 2+ branch of ^{207}Bi, and the results shown for ^{207}Bi do not include this contribution. Also, with regard to both the SRP and SL theories, all calculations for forbidden decays have been performed without the inclusion of shape factors. However, this is not expected to affect the results significantly since, in the normal approximation, they are predicted to be unity for all cases under consideration.

The binding energies which were used in obtaining the SRP results (those listed in Table I) were taken from stand-

TABLE I. K-shell internal ionization probabilities in EC decay.

Nuclide	E (keV)	$B_K(i)$ (keV)	$B_K(f)$ (keV)	σ	$P_K(\times 10^{-5})$ Present work Unscreened	Screened	SL
^{37}Ar	813.8	3.20	2.82	0.579	20.52	25.84	52.94
^{54}Mn	542.2	6.54	5.99	0.620	9.41	11.25	11.36
^{55}Fe	231.4	7.11	6.54	0.624	7.90	7.42	20.06
^{71}Ge	235.7	11.10	10.37	0.644	4.38	5.08	11.84
^{85}Sr	550.0	16.10	15.20	0.663	2.98	3.38	9.38
^{103}Pd	506.3	24.35	23.22	0.682	1.56	1.74	6.03
^{109}Cd	94.3	26.71	25.51	0.685	0.30	0.34	0.89
^{113}Sn	647.1	29.20	27.94	0.689	1.21	1.23	5.33
^{131}Cs	355.0	35.98	34.56	0.699	0.68	0.75	3.22
^{165}Er	377.1	57.49	55.62	0.716	0.24	0.26	1.71
^{181}W	188.0	69.53	67.42	0.722	0.020	0.022	0.14
^{207}Bi	771.7	90.53	88.00	0.731	0.10	0.11	1.97

ard references and are essentially exact. This implies that, to a large extent, the influence of the other orbital electrons on the two initial K electrons has been taken into account. The explanation for this is that, in the vicinity of the K shell, the potential due to all the other electrons, although large, is fairly constant, and is well represented by[11] $V(r)=A+B(r)$, with $B(0)=0$ and $B(r)\ll A$. The constant term has no effect on the initial-state wave function; it simply contributes to the binding energy, and it is the second term $B(r)$ whose effects are simulated by the introduction of the screening parameter. Thus, the use of exact binding energies together with a screening parameter ought to simulate the effects of $V(r)$ quite well. Unfortunately, the effects of the orbital electrons on the final state cannot be incorporated so easily.

Finally, in Table II we have retabulated the screened SRP and SL results and compared them with all recent experimental data.[28] Arbitrarily, we have included only those measurements that have been published since 1970. A summary of earlier experimental work may be found in Ref. 5. For completeness, we have also included the predictions of the Primakoff-Porter (PP) theory[1] and, where available, results of the work of Mukoyama *et al.* (MIKS).[29] These latter authors also used the variational approach but, in their evaluation of (3), they employed relativistic hydrogenic wave functions that were shielded, with effective charges extracted from a relativistic SCF calculation. But because of the sensitivity of these results to the values of the screening constants, the MIKS results are not expected to be as reliable as the SL results.

It is evident that the SRP results are systematically larger than those of MIKS by about 5%–10% for all except large Z nuclides (for which the SRP results are not expected to be too accurate) and therefore stand slightly closer to the experimental values. In contrast, the SL results are anywhere from 1.5–5 times larger than the SRP results. Indeed, these two sets of predictions bracket the experimental values for the majority of the nuclides studied, these data lying somewhat closer to the SRP predictions. On the other hand, for ^{65}Zn, ^{71}Ge, and ^{109}Cd the experimental values are somewhat larger than the SL predictions, but in reasonable agreement with them. However, for ^{181}W there is serious disagreement with both theories.

What is perhaps the most striking feature of Table II is the remarkably good agreement with experiment which is achieved by the Primakoff-Porter results. Their theory is based upon a completely nonrelativistic treatment using a Coulomb wave function for the final state and, for the initial state, a correlated wave function of the form

$$\psi(\mathbf{r}_1,\mathbf{r}_2)\propto e^{-\gamma_1(r_1+r_2)}e^{\gamma_2|r_1-r_2|}\ ,$$

with the parameters γ_1 and γ_2 chosen to make the wave function a good fit to the Hylleraas variational wave function for the two-electron ion. Considering the simplicity of their approach and their neglect of relativistic effects and the influence of the other orbital electrons, one can only regard this good agreement as fortuitous.

Clearly, there is a need for further theoretical work for the purpose of reconciling the differences between the SRP and SL theories. Each approach has its strengths and weaknesses. The SL theory is fully relativistic and takes good account of the influence of the other orbital electrons. (This is particularly important for the final state.) However, because it is based on an independent particle model, the SL approach does not adequately take into account the correlation between the two K electrons in the initial state. In contrast, the SRP theory does treat well the initial-state correlation, but it is not fully relativistic and does not recognize

TABLE II. Comparison of various theories with recent experimental results.

	$P_K(\times10^{-5})$					
	Theory					
Nuclide	PP	MIKS	SRP	SL	Expt.	Ref.
^{54}Mn	13.53		11.25	24.3	36 ±3	12
^{55}Fe	11.48	8.81	9.42	20.06	12 ±4	13
					10.1 ±2.7	14
^{65}Zn	8.65			15.3	22 ±2	15
^{71}Ge	6.92	4.56	5.08	11.84	12	16
^{85}Sr	5.27		3.38	9.38	6.0 ±0.5	17
^{103}Pd	3.27		1.74	6.03	3.13 ±0.31	18
^{109}Cd	0.63		0.34	0.89	15.2 ±2.4	19
					2.8 ±0.7	20
					1.02 ±0.36	21
^{113}Sn	2.79		1.34	5.33	1.5 ±0.5	22
^{131}Cs	1.76	0.71	0.75	3.22	1.33 ±0.33	23
					2.3 ±0.3	24
					1.4 ±0.1	25
^{165}Er	0.87	0.30	0.26	1.71	0.67 ±0.39	23
					0.82 ±0.28	26
^{181}W	0.076		0.022	0.14	0.24 ±0.06	26
					1.25 ±0.42	22
^{207}Bi	0.69		0.11[a]	1.97	0.6 ±0.25	27

[a]Contribution from 10% 2^+ branch not included.

the influence of the orbital electrons on the scattering state, a serious defect.

It appears that what is needed is a hybrid approach which combines the advantages of the SL and SRP theories while eliminating their weaknesses. Indeed, such an approach is currently under investigation. In addition, there is a need for more high-precision experiments on several of the nuclides listed, in the hope of further clarifying the extent of the discrepancy between theory and experiment.

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