PHYSICAL REVIEW C VOLUME 19, NUMBER 3 MARCH 1979

Atomic electron excitation probabilities during orbital electron capture by the nucleus

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(Received 6 September 1978)

Approximate probabilities of electron excitation (shakeup/shakeoff) from various atomic states during nuclear *ns* electron capture have been calculated in the sudden approximation, using Hartree-Fock wave functions. Total excitation probabilities are much lower than during inner-shell ionization by photons or electrons, and *ns* states are more likely to be excited than *np* states. This latter result is borne out by $K\alpha$ x-ray satellite spectra.

[RADIOACTIVITY Atomic electron excitation (shakeup/shakeoff) during nuclear electron capture, calculated form Hartree-Fock wave functions.]

I. INTRODUCTION

Radioactive decay by nuclear electron capture can be accompanied, with low probability, by the excitation of orbital electrons to unoccupied bound states or to the continuum.[1] This process can be treated as "shakeup/shakeoff" ("internal excitation/ionization") caused by the change in the Coulomb potential during the decay. The potential varies suddenly, on an atomic time scale, as the neutral parent atom of atomic number Z turns into a daughter ion of atomic number $Z'=Z-1$ with an inner-shell vacancy left by the captured electron. Atomic-electron screening of the nuclear charge is decreased by formation of the inner-shell hole, but the reduction in screening does not fully compensate for the decrease in nuclear charge by one unit. The opposing effects of reduced screening and reduced nuclear charge do, however, cause the production of a second inner-shell vacancy during electron capture to be much less probable than shakeup/shakeoff during beta decay[2,3] or photoionization (or ionization by electron impact).[4,5] Another striking difference between electron excitation accompanying nuclear electron capture and during electron-impact ionization exists with regard to the specific atomic subshells in which the effect predominantly occurs; this difference was recently discovered in a study of $K\alpha$ x-ray satellites.[6] The present work was undertaken to examine the theoretical basis for these differences.

II. THEORY

The probability of electron ejection during nuclear electron capture is

$$dw = 2\pi \int \frac{1}{2} \sum |M|^2 \delta(W_0 + 1 - |E'_{nl}| - W - q) d\vec{q} d\vec{p}\,, \tag{1}$$

where $\vec{p}$ and W are the momentum and total relativistic energy of the ejected electron, $\vec{q}$ is the neutrino momentum, q is its energy, $1-|E'_{nl}|$ is the total energy of an nl electron in the daughter atom (with binding energy E'_{nl}), and W_0+1 is the energy difference between the parent atom and the neutral daughter atom.[1] The units are such that $\hbar = m = c = 1$. The summation in Eq. (1) extends over spin states of ejected electron and neutrino, and of the initial nl electrons. Furthermore, one must sum over final and average over initial nuclear spin states. For the case of K capture, these operations have been discussed by Intemann.[7] An analogous expression can be written for electron excitation to bound states.

The most difficult aspect of the quantitative evaluation of Eq. (1) is a proper formulation of the matrix element $|M|$, which contains the overlap of the ejected-electron continuum wave function in the potential of the daughter ion with that electron's bound-state wave function in the parent atom. Understandably, the effect of electron-electron Coulomb correlation on the magnitude of the matrix element is strong; this adds to the difficulty of formulating $|M|$.[1,8-10] The problem of calculating a matrix element that involves the continuum wave function can, however, be circumvented if one is only interested in an approximate expression for the total probability of electron excitation, and not in the energy spectrum of the ejected electrons.

III. APPROXIMATE ESTIMATE OF THE PROBABILITY OF ELECTRON EXCITATION DURING NUCLEAR ELECTRON CAPTURE

For many practical purposes it is desirable to have an approximate theoretical estimate of the probability of electron excitation from various atomic shells during nuclear K-, L-, ... electron capture. Such an estimate can be made quite simply by using the fact that, in the sudden approximation, the amplitude of the probability that an atomic electron retains its original quantum numbers is proportional to the overlap of its original wave function with its wave function in the daughter ion:

$$P_{\text{remain}} \propto \left| \int \psi_{nl}'^{*}(\vec{r})\psi_{nl}(\vec{r})d\vec{r} \right|^2 . \tag{2}$$

In the same manner in which Carlson *et al.* have made estimates of shakeup/shakeoff probabilities during photoionization and beta decay,[3,5] we can therefore write an approximate expression for the probability that an nl electron is promoted to a higher bound state or to the continuum during electron capture,

$$P = 1 - \left[\left| \int \psi_{nl}'^{*}(\vec{r})\psi_{nl}(\vec{r})d\vec{r} \right|^2 \right]^N - P_0 . \tag{3}$$

Here, subtraction of P_0 excludes transitions to occupied bound states, forbidden by the Pauli principle,

$$P_0 = \sum_{n' \neq n}^{n'_{\max}} \frac{NN'}{2(2l+1)} \left| \int \psi_{n'l}(\vec{r})\psi_{nl}(\vec{r})d\vec{r} \right|^2 , \tag{4}$$

where $N(N')$ is the number of electrons in the nl $(n'l)$ shell, and ψ' and ψ are the final and initial wave functions, respectively. The summation extends over all occupied $n'l$ states. A change in the orbital angular-momentum quantum number l during the transition is forbidden by monopole selection rules.

IV. RESULTS AND DISCUSSION

The probability of electron excitation (shakeup/shakeoff) from the various subshells during nuclear ns-electron capture was calculated according to Eq. (3) with the Froese-Fischer Hartree-Fock program.[11] Results for a number of elements with $7 \leq Z \leq 54$ are listed in Table I. As the probabilities vary smoothly with atomic number, they can be interpolated easily for elements between those listed. These calculations are nonrelativistic, hence they were not extended above $Z = 54$.

It is interesting to compare the probabilities of electron excitation during nuclear electron capture with those for photoionization (or electron-impact ionization). For the latter, we refer to the tables of Carlson and Nestor[5]; results from the present calculation for the case of photoionization coincide with those of Ref. 5 within the accuracy of the model, and hence are not repeated here.

A typical case is illustrated in Fig. 1, where we compare electron excitation during K capture and K-shell photoionization of Kr. The total excitation probability is seen to be over two orders of magnitude lower during electron capture than during photoionization (0.171% vs 21.5%); only K electrons are more likely to be excited during capture (0.016% vs <0.005%). In capture, ns electrons are several times as likely to be excited than np electrons, while the opposite holds for photoionization.

This fact is borne out experimentally by the observation that $K\alpha_{3,4}$ x-ray satellites, which arise from $[1s][2p]$ vacancy states and appear prominently in electron-impact excited spectra,[12] are virtually undetectable in x-ray spectra from radioactive ^{55}Fe and ^{71}Ge sources.[6] In the K x-ray spectra from electron-capture sources, on the other hand, the $K\alpha_3''$ satellites stand out, which arise

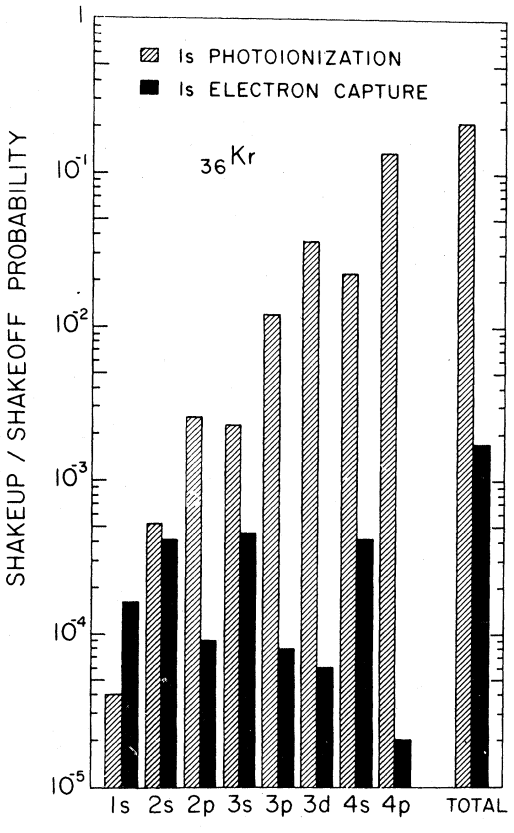

FIG. 1. Probabilities of electron excitation from various nl states during K-electron photoionization and during K-electron capture of Kr, calculated in the sudden approximation according to Eq. (3).

TABLE I. Electron excitation probabilities (in percent) from various atomic subshells during nuclear electron capture (EC).

Z	Element	EC from subshell	Probability (%) of electron excitation from subshell 1s	2s	2p	3s	3p	3d	4s	4p	4d	5s	Total probability
7	N	1s	0.655	1.917	0.042								2.614
		2s	3.145	3.326	9.626								16.096
10	Ne	1s	0.297	0.955	0.029								1.280
		2s	1.390	1.423	7.289								10.102
12	Mg	1s	0.193	0.548	0.030	0.279							1.049
		2s	0.898	0.914	4.575	1.274							7.661
		3s	0.910	3.527	11.760	9.505							25.703
15	P	1s	0.115	0.308	0.032	0.246	0.007						0.708
		2s	0.529	0.525	2.407	1.026	0.365						4.852
		3s	0.536	1.839	5.409	2.110	9.104						18.999
18	Ar	1s	0.076	0.201	0.027	0.181	0.003						0.489
		2s	0.347	0.340	1.343	0.731	0.629						3.389
		3s	0.351	1.137	2.818	1.130	8.210						13.646
20	Ca	1s	0.060	0.153	0.023	0.129	0.005	0.083					0.452
		2s	0.271	0.255	0.976	0.548	0.646	0.243					2.939
		3s	0.274	0.832	1.971	0.787	5.790	1.037					10.691
23	V	1s	0.044	0.112	0.019	0.113	0.010	0.051	0.108				0.458
		2s	0.197	0.185	0.688	0.490	0.708	0.316	0.392				2.977
		3s	0.199	0.590	1.331	0.545	3.830	0.781	0.679				7.956
		4s	0.200	0.590	1.375	1.650	5.842	14.489	3.124				27.271
26	Fe	1s	0.033	0.087	0.017	0.096	0.013	0.037	0.097				0.380
		2s	0.150	0.142	0.514	0.410	0.621	0.189	0.356				2.382
		3s	0.152	0.444	0.968	0.404	2.740	1.167	0.466				6.341
		4s	0.152	0.441	0.992	1.157	3.912	12.482	2.922				22.058
30	Zn	1s	0.024	0.064	0.013	0.078	0.013	0.024	0.081				0.298
		2s	0.110	0.104	0.370	0.323	0.498	0.106	0.292				1.803
		3s	0.111	0.322	0.678	0.289	1.898	1.355	0.307				4.959
		4s	0.111	0.317	0.691	0.796	2.584	11.018	2.784				18.303
32	Ge	1s	0.021	0.055	0.011	0.063	0.011	0.014	0.062	0.001			0.238
		2s	0.095	0.089	0.313	0.258	0.401	0.044	0.198	0.039			1.438
		3s	0.096	0.273	0.567	0.240	1.566	1.343	0.347	0.094			4.525
		4s	0.096	0.270	0.581	0.707	2.403	6.506	1.114	5.188			16.864
36	Kr	1s	0.016	0.042	0.009	0.045	0.008	0.006	0.043	0.002			0.171
		2s	0.073	0.067	0.226	0.183	0.269	0.018	0.137	0.107			1.081
		3s	0.074	0.202	0.401	0.176	1.016	1.124	0.317	0.400			3.710
		4s	0.074	0.200	0.413	0.530	1.608	3.558	0.586	5.419			12.389
40	Zr	1s	0.013	0.033	0.008	0.033	0.006	0.004	0.031	0.003	0.014	0.035	0.179
		2s	0.058	0.051	0.169	0.133	0.199	0.014	0.110	0.135	0.069	0.101	1.038
		3s	0.059	0.153	0.296	0.129	0.721	0.878	0.266	0.507	0.029	0.160	3.197
		4s	0.059	0.152	0.305	0.378	1.111	2.288	0.359	3.080	0.623	0.469	8.824
44	Ru	1s	0.011	0.026	0.006	0.026	0.005	0.002	0.028	0.004	0.010	0.037	0.153
		2s	0.047	0.041	0.132	0.105	0.158	0.016	0.099	0.134	0.038	0.107	0.877
		3s	0.047	0.121	0.228	0.102	0.557	0.645	0.237	0.477	0.072	0.146	2.633
		4s	0.048	0.120	0.236	0.295	0.841	1.470	0.266	2.118	1.446	0.274	7.113
48	Cd	1s	0.009	0.021	0.005	0.021	0.005	0.001	0.024	0.004	0.008	0.033	0.131
		2s	0.039	0.033	0.105	0.086	0.131	0.017	0.089	0.123	0.021	0.096	0.739
		3s	0.039	0.098	0.181	0.084	0.446	0.463	0.207	0.423	0.141	0.122	2.201
		4s	0.039	0.097	0.187	0.237	0.662	0.953	0.209	1.586	1.825	0.183	5.980

TABLE I. *(Continued)*.

Z	Element	EC from subshell	1s	2s	2p	3s	3p	3d	4s	4p	4d	5s	5p	Total probability
51	Sb	1s	0.008	0.018	0.005	0.018	0.004	0.001	0.020	0.003	0.004	0.023	0.000	0.103
		2s	0.034	0.028	0.090	0.073	0.111	0.016	0.072	0.102	0.008	0.062	0.021	0.618
		3s	0.034	0.084	0.153	0.071	0.371	0.376	0.172	0.365	0.175	0.103	0.053	1.955
		4s	0.035	0.083	0.158	0.198	0.541	0.735	0.171	1.244	1.723	0.244	0.155	5.286
54	Xe	1s	0.007	0.016	0.004	0.016	0.003	0.000	0.017	0.003	0.004	0.018	0.001	0.087
		2s	0.030	0.025	0.077	0.063	0.095	0.016	0.061	0.084	0.006	0.051	0.040	0.546
		3s	0.030	0.073	0.131	0.060	0.307	0.311	0.148	0.305	0.193	0.094	0.117	1.770
		4s	0.030	0.072	0.135	0.167	0.443	0.584	0.144	1.157	1.534	0.243	0.379	4.889

from [1s][2s] hole states (Fig. 2). The relative intensities of the satellites in the two cases agree qualitatively with the calculated 2s/2p excitation-probability ratios (Table II).

The fact that the 2s hole should remain as a "spectator hole" during radiative decay of the 1s hole seems somewhat surprising, because the calculated 2s level width in a singly ionized Mn or Ga atom is larger than the 1s width.[13-15] There are, however, considerable uncertainties associated with these calculations. Furthermore, the simultaneous presence of a 1s vacancy in the doubly ionized atom can be expected to raise the 2s binding energy so as to cut off some of the intense Coster-Kronig transitions that govern the 2s width, thus increasing the lifetime of the 2s vacancy.

In conclusion, it should be noted that use of Eq. (3) implies that the sudden approximation of perturbation theory is applicable, and that the energy-conserving delta function in Eq. (1) can be neglected. These assumptions are reasonably good in the limit of large decay energy ($q \gg |E'_{nl}|$). Nevertheless, the simple overlap approach of Eq. (3) is exceedingly sensitive to the fine details of the wave functions, and good accuracy cannot be expected even with Hartree-Fock wave functions. A further difficulty arises from the use of any independent-particle model, including Hartree-Fock, to compute an effect that is the very epitome of correlation, viz., shakeup/shakeoff. The present calculations and those of Ref. 5 can presumably be relied upon for an approximate comparison of relative electron excitation probabilities in photoionization vs electron capture, but the absolute probabilities are perhaps no better than order-of-magnitude estimates. Thus the present calculations predict a double K-shell ionization probability during electron capture of 33×10^{-5} for $_{26}$Fe; a more sophisticated (shakeoff only) calculation by Intemann[16] yields a probability of 8×10^{-5}, and Mukoyama *et al.*[17] calculate 16×10^{-5} for the shakeoff probability. A measurement by Charpak[18] showed that the double K-vacancy production is $(38\pm17)\times10^{-5}$. There is an obvious need for additional theoretical and experimental work on this interesting subject.

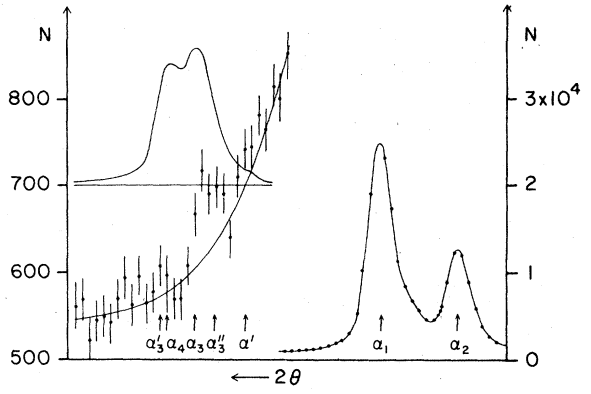

FIG. 2. $K\alpha$ satellites (left) and diagram lines (right) from ^{55}Fe electron-capture decay, compared with Parratt's Mn $K\alpha$ satellite spectrum produced by electron-impact ionization (upper left, same scale) (Ref. 12). In electron capture, the $K\alpha'_3$ satellite stands out, produced by [1s][2s] double ionization, while in electron-impact ionization the $K\alpha_{3,4}$ satellites from [1s][2p] double ionization prevail.

TABLE II. Probability of 2s- and 2p-electron excitation during K electron capture and K-shell ionization.

Element	Ionization mechanism	Excitation probability (%) 2s	2p	Ratio 2s/2p
$_{26}$Fe	K–EC	0.087	0.017	5.1
$_{25}$Mn	K ionization	0.126	0.692	0.2
$_{32}$Ge	K–EC	0.055	0.011	5.0
$_{31}$Ga	K ionization	0.077	0.404	0.2

ACKNOWLEDGMENTS

One of us (B.C.) gratefully acknowledges the hospitality extended to him, during the summer of 1977, at the Laboratoire de Physique Atomique et Nucléaire, Institut du Radium. This work was supported in part by the National Aeronautics and Space Administration (Grant No. NGR 38-003-036) and by the U. S. Army Research Office (Grant No. DAAG 29-78-G-0010).

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