

Electron Shakeoff Accompanying Photoionization

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Received February 4, 1986

The electron shakeoff probabilities accompanying K-shell photoionization have been calculated relativistically using the screened hydrogenic wave functions. The screening constants were estimated by adjusting the mean radius with that obtained with the relativistic Hartree-Fock-Slater model. Calculations were made in one-step approach and the shakeoff probabilities were obtained as a function of incident photon energy. It is found that for the case of K-electron ejection the energy dependence of the probability is appreciable. The calculated values are compared with other theoretical calculations and the experimental results.

KEY WORDS: Electron Shakeoff/ Photoionization/ Screened Hydrogenic Model/

I. INTRODUCTION

As the result of photoionization, the atomic electrons experience a sudden change in central potential due to removal of an electron and have a small probability to be ejected into the continuum. This process, called *electron shakeoff*, is one of the main sources of the multiple ionization in photoionization and important in x-ray photoelectron spectroscopy.

Carlson and Nestor¹⁾ made theoretical calculations on electron shakeoff probabilities as the result of photoionization of the rare gases in the sudden approximation, using the relativistic Hartree-Fock-Slater (RFHS) wave functions. Their model is based on the two-step approach and probability is independent of the incident energy of x rays. Such a model is valid only when the kinetic energy of photoelectron ejected is much larger than that of the shakeoff electron.

In addition, it is well known that the sudden approximation with the single-electron wave functions, such as the Hartree-Fock (HF) and Hartree-Fock-Slater (HFS) wave functions, agree with experiment so long as the photoelectric effect takes place from a shell that is inside the one from which the shakeoff electron is ejected. When a sudden creation of a vacancy in a given shell promotes additional electron ejection from the same shell, their model underestimates the shakeoff probabilities.²⁾

On the other hand, for double photoionization cross sections of He atom, the inclusion of electron correlation in the initial ground-state wave function was found to be very important.³⁻⁵⁾ These calculations used the Hylleraas-type wave functions which include the correlation effect explicitly and give a reasonable agreement with the experimental data. However, they are nonrelativistic and limited only for He.

Recently we have developed the screened hydrogenic model for the electron shakeoff processes accompanying electron capture⁶⁻⁹⁾ and internal conversion.^{10,11)}

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The calculated shakeoff probabilities are in fairly good agreement with the recent experimental results, even in the case where the shakeoff electron is ejected from the same shell as the one in which the initial vacancy is produced, i.e. K-shell electron ejection during K-electron capture or K-shell conversion. In the two-step approach, the electron shakeoff probabilities accompanying internal conversion¹⁰⁾ are equivalent to those accompanying photoionization. Agreement between theory and experiment for internal conversion indicates that the screened hydrogenic model is also valid for electron shakeoff accompanying photoionization by photons with high incident energies. However, in order to obtain more refined values for the electron shakeoff probability during photoionization, the one-step approach should be used, in which two electrons (a photoelectron and a shakeoff electron) are considered to be ejected simultaneously.

In the present work, we have calculated the K- and L-electron shakeoff probabilities accompanying the K-shell photoionization in the one-step approach. Calculations have been made using screened hydrogenic wave functions with screening constants estimated from the RHFS model.

II. THEORETICAL MODEL

As has been made in the case of internal conversion,¹¹⁾ we have introduced some assumptions to simplify the problem. First, we neglect the effect of antisymmetrization between two electrons in the final state, a shakeoff electron and a photoelectron. It is well known that the energy distribution of the shakeoff electrons is concentrated in the low-energy region,¹⁰⁾ while the photoelectron has an energy near to the normal photoionization line, forming a broad satellite peak. Considering this fact, we assume the effect of electron exchange in the final state to be small, except for the case where the energy of the incident photon is very close to the threshold of photoionization.

Second, we ignore the contribution from the direct collision process. This process is Coulomb scattering of the atomic electron by the photoelectron and considered as an alternative ionization mechanism accompanying photoionization. The probability of direct collision has been estimated in the case of nuclear β decay by Feinberg.¹²⁾ His results indicate that the relative probability of direct collision to shakeoff is small when the ratio of the binding energy of the atomic electron to the kinetic energy of the photoelectron is small.

Third, the K-electron ejection following the outer-shell photoionization is neglected. This process cannot be distinguished experimentally from the outer-shell electron shakeoff accompanying K-shell photoelectric effect because the final state of the atom is same. The experimental data should include both contributions. However, the theoretical calculations of Carlson and Nestor¹³⁾ show that the probability of the K-electron shakeoff accompanying the outer-shell photoelectric effect is smaller than its counterpart.

Finally, the contribution from the shakeup process is not taken into account. The sudden change in the central potential causes also electron excitation to an unoccupied bound state. For outer-shell electrons, the probability of this process may be large. However, in the case of K- and L-shell electrons the shakeup probability is

considered to be small, except for very light elements, since the outer shells near to these shells are already occupied.

According to the theory of photoionization, the cross section of K-shell photoelectric effect is given by¹³⁾

$$\sigma_K = \frac{3Z^5\alpha^4}{2k^5} \phi_0 (\gamma^2 - 1)^{3/2} \left\{ \frac{4}{3} + \frac{\gamma(\gamma - 2)}{\gamma + 1} \left[1 - \frac{1}{2\gamma(\gamma^2 - 1)^{1/2}} \ln \frac{\gamma + (\gamma^2 - 1)^{1/2}}{\gamma - (\gamma^2 - 1)^{1/2}} \right] \right\}, \quad (1)$$

where $\gamma = (1 - \beta^2)^{-1/2}$, $\beta = p/W_0$, $\phi_0 = \frac{8}{3}\pi\alpha^2$, Z is the atomic number, a is the fine structure constant, k is the energy of the incident photon, and p and W_0 are the momentum and the total energy of the ejected electron. Denoting the binding energy of the K-shell electron as B_K , the total energy of the photoelectron in the ordinary photoelectric effect is

$$W_0 = k + 1 - B_K. \quad (2)$$

When an i -shell electron makes a transition from an initial state ψ_{2i} to a final state ψ_{2f} during K-shell photoionization the cross section is given by

$$\sigma_{Ki} dW_2 = \frac{1}{2\pi^2} |\langle \psi_{2f} | \psi_{2i} \rangle|^2 \sigma_K(W_1) p_2 W_2 dW_2, \quad (3)$$

where p_2 and W_2 are the momentum and total energy of the shakeoff electron and $\sigma_K(W_1)$ is the cross section of the K-shell photoelectric effect for the incident photon energy k and the total energy of the photoelectron W_1 . The energy relation corresponding to Eq. (3) is

$$W_1 + W_2 = k + 2 - B_K - B_i. \quad (4)$$

Here B_i is the binding energy of the i -shell electron before ejection.

From Eqs. (1) and (3), the differential shakeoff probability of the i -shell electron accompanying K-shell photoionization is obtained as a function of W_2 :

$$P_{Ki}(W_2) dW_2 = \frac{n_i}{2\pi^2} |\langle \psi_{2f} | \psi_{2i} \rangle|^2 \frac{\sigma_K(W_1)}{\sigma_K(W_0)} p_2 W_2 dW_2, \quad (5)$$

where n_i is the number of electrons in the i -shell. It should be noted that in the case of K-electron shakeoff, n_i is taken to be unity, because only one electron is available for the shakeoff process.

The total i -shell shakeoff probability per K-shell photoionization is calculated by integrating Eq. (5) over the electron energy W_2 :

$$P_{Ki} = \int_1^{W_{\max}} P_{Ki}(W_2) dW_2, \quad (6)$$

where the maximum energy available for the shakeoff electron is

$$W_{\max} = k + 1 - B_K - B_i. \quad (7)$$

The relevant expressions of the atomic matrix element, $\langle \psi_{2f} | \psi_{2i} \rangle$, for ejection of K- and L-shell electrons were already derived by us using the relativistic hydrogenic

wave functions in the Coulomb field [Eqs. (5) and (6) in Ref. 10]. In the practical calculations, the effective nuclear charge was used instead of the atomic number Z . The method to obtain the screening constant is discussed later.

The photoionization cross section as a function of the ejected electron energy in Eq. (5), $\sigma_K(W_1)$, can be expressed as the same form as the ordinary photoionization cross section, Eq. (1). The ejected electron energy W_0 in Eq. (1) is replaced by W_1 for the fixed value of k .

In order to take into account the Coulomb interaction between atomic electrons, a screening constant is used as in our previous works,⁶⁻¹¹ and the nuclear charge Z in the atomic matrix element is replaced by the effective one $Z_{\text{eff}} = Z - \sigma$, where σ is the screening constant.

For the initial bound state, σ is determined from the relation¹⁴⁾

$$\sigma = Z(1 - \bar{r}_Z / \bar{r}_{\text{SCF}}), \quad (8)$$

where \bar{r}_Z is the mean relativistic hydrogenic radius and \bar{r}_{SCF} is the mean radial distance calculated with the RHFS wave functions.¹⁵⁾

In the case of continuum electrons, the screening constant is taken as the same value as that for the bound electron to be ejected. This choice is justified from the fact that the largest contributions to both the atomic matrix element and the photoionization cross section come from the region near to the mean radius of the bound electron concerned.

In the present model, two electrons are considered to be emitted simultaneously. However, the mechanisms for ejection of these electrons are different. The photoelectron is ejected due to electromagnetic interaction between the photon and the electron, while the emission of the shakeoff electron arises from the sudden change in the central potential. Considering the different mechanisms for these two electrons, we use the different screening constants for the final continuum electrons. The final-state screening constant for the shakeoff electron should be determined by taking into consideration of the existence of the K-shell vacancy produced by photoionization.

The screening constant for the photoelectron is chosen as the same as that for the bound-state electron, as described above. On the other hand, the screening constant for the shakeoff electron in the final continuum state is obtained by

$$\sigma_C = (\sigma_h / \sigma_s) \sigma. \quad (9)$$

Here σ is the screening constant determined from Eq. (3), and σ_h and σ_s are the Slater's screening constants for the atom with a hole in the K shell and for the ordinary atom, respectively.¹⁶⁾

The explicit expression for \bar{r}_Z is given by us [Eq. (12) in Ref. 10]. For the values of \bar{r}_{SCF} , we used the table prepared by Carlson et al.¹⁵⁾

In order to compare with the present model in the one-step approach, the shakeoff probability in the two-step model is also calculated within the framework of the screened relativistic hydrogenic model. This probability is equal to the high-energy limit of Eq. (5) and can be given by a simple wave-function overlap:¹⁰⁾

$$P_{Ki}^0(W_2) dW_2 = \frac{n_i}{2\pi^2} |\langle \psi_{2f} | \psi_{2i} \rangle|^2 p_2 W_2 dW_2. \quad (10)$$

The same screening constants described above are used in Eq. (10).

III. RESULTS AND DISCUSSION

We have calculated the K- and L-shell electron shakeoff probabilities per K-shell photoionization for several elements. The probabilities are obtained as a function of the energy of the incident photon. The binding energies were taken from the table of Bearden and Burr.¹⁷⁾ All the numerical calculations of the present work have been performed on the FACOM M-382 computer of the Data Processing Center of Kyoto University.

In Fig. 1, the K-shell shakeoff probability accompanying K-shell photoionization in Ne is plotted as a function of incident photon energy. The photon energy is expressed in terms of the reduced energy, i.e. a ratio to the threshold energy of the shakeoff process, which in this case corresponds to twice of the K-shell binding energy of Ne atom.

The solid curve represents the present results and the dashed curve indicates the two-step model with the screened hydrogenic wave functions. For high-energy photons, the one-step model approaches to the two-step results. The dot-dashed line is the calculated results of Carlson and Nestor.¹⁾ As has been described above, their model gives considerably smaller values than the present model.

In order to see the effect of choice of the screening constants, the calculations with

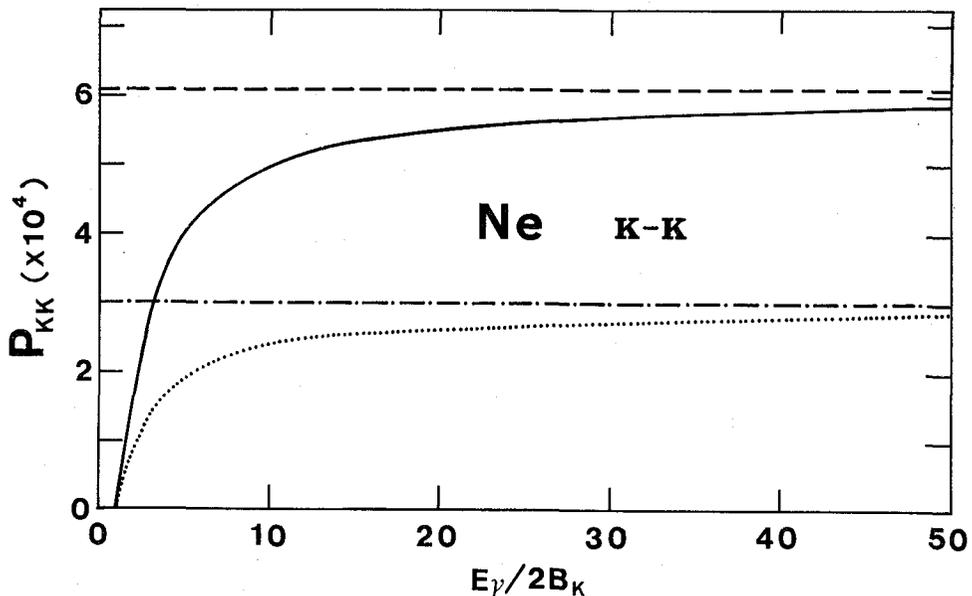


Fig. 1. K-shell shakeoff probability accompanying K-shell photoionization in Ne as a function of the reduced energy. The solid curve has been calculated from the present theory with relativistic screening constants; the dashed curve from the two-step model; the dotted curve from the present theory with Slater's screening constants; the dot-dashed curve from the two-step calculation of Carlson and Nestor (Ref. 1).

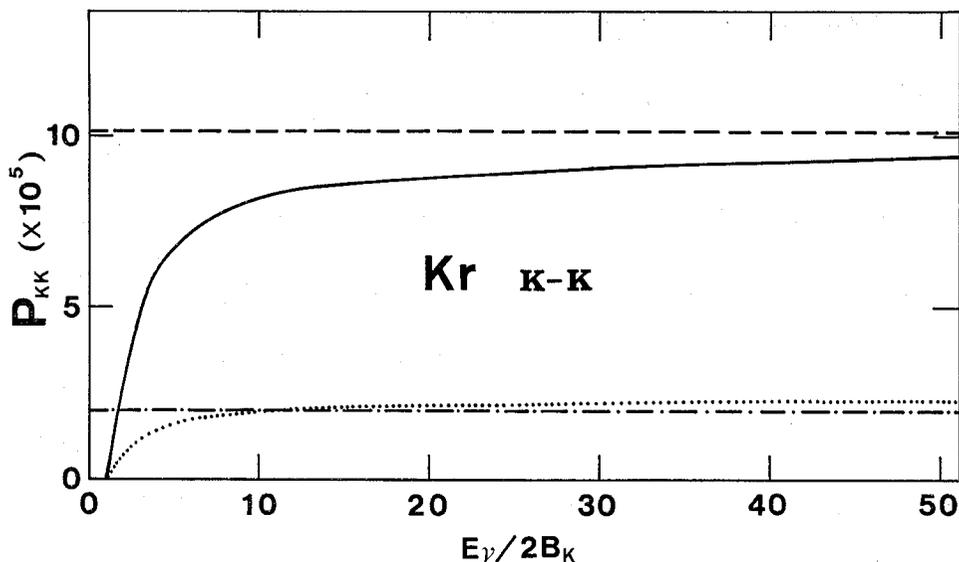


Fig. 2. K-shell shakeoff probability accompanying K-shell photoionization in Kr as a function of the reduced energy. See caption of Fig. 1.

the Slater's screening constants¹⁰⁾ have also been made. The results are shown in the figure by the dotted curve. It is clear that the use of the relativistic screening constants increases the shakeoff probability. This fact has already been shown by us for the case of the ejected shakeoff electron spectrum during internal conversion.¹⁰⁾ In addition, it is interesting to note that in high-energy limit the results with the Slater's screening constants are close to the two-step calculations of Carlson and Nestor.¹⁾

Figure 2 shows the K-shell shakeoff probability during K-shell photoionization of Kr atom. The general trend of the energy dependence of the shakeoff probability is quite similar to the case of Ne. For Kr, the relativistic calculations with the Slater's screening constants are nearly equal to the results of Carlson and Nestor in the high-energy limit.

In Fig. 3, the similar results for the L_1 -shell electron shakeoff probability in K-shell photoelectric effect in Kr is plotted as a function of the reduced energy of the incident photon. In this case, the reduced energy is defined as the ratio of the photon energy to the sum of the K- and L_1 -shell binding energies of Kr. For L_1 shell the curve with the Slater's screening constants is slightly higher than that with the relativistic screening constants. The two-step model of Carlson and Nestor is also in agreement with the present values. All four curves are close each other for high-energy photons.

Comparison of energy dependence of the K-, L_1 -, and L_2 -shell shakeoff probabilities is shown in Fig. 4. The L_3 -shell curve almost coincides with the L_2 -shell one and is not plotted in the figure. The photon energy is given in the form of the reduced energy, while the shakeoff probability is expressed as the ratio to the corresponding value in the two-step model, P_{Ki}/P_{Ki}^0 . It can be seen from the figure that the shakeoff probability increases steeply with increasing photon energy and reaches to the value in the two-step model. At high energies, the probability is almost constant, i.e.

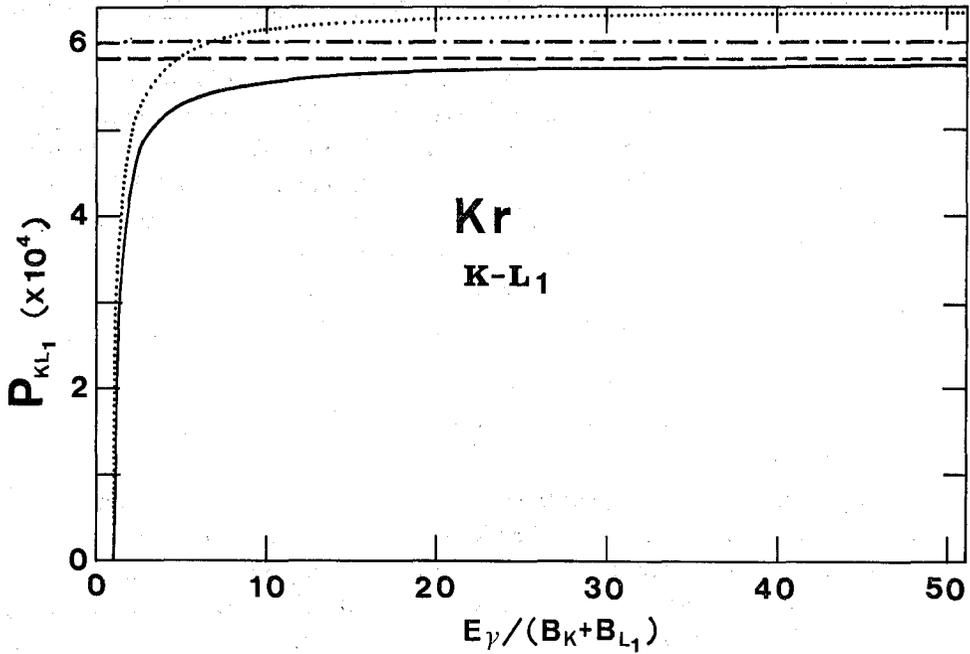


Fig. 3. L₁-shell shakeoff probability accompanying K-shell photoionization in Kr as a function of the reduced energy. See caption of Fig. 1.

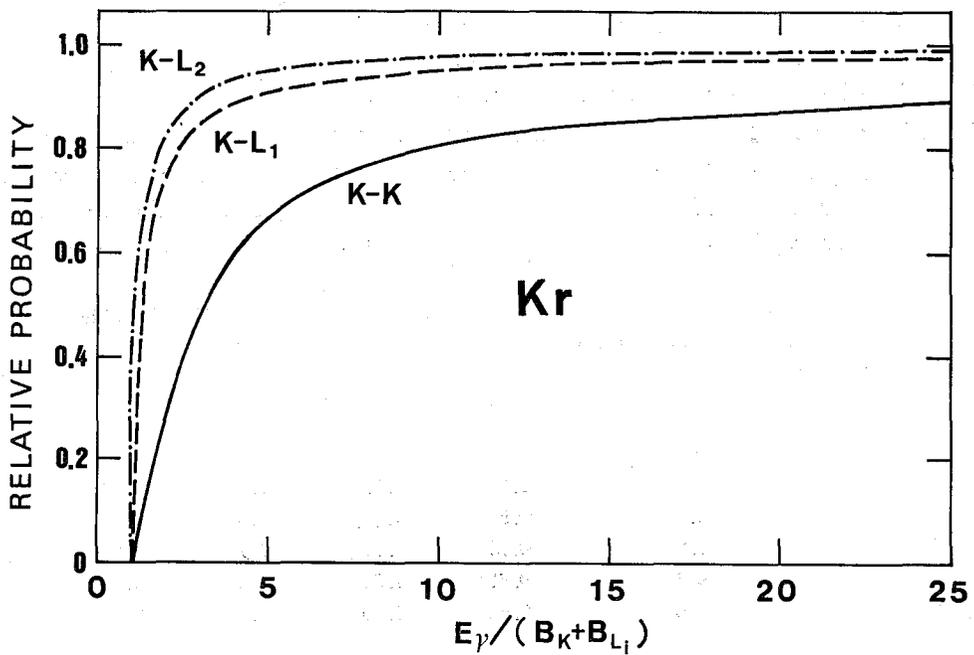


Fig. 4. Comparison of K-, L₁-, and L₂-shell shakeoff probabilities accompanying K-shell photoionization in Kr as a function of the reduced energy. The probability is expressed as a ratio to the value obtained from the two-step model.

independent of the incident photon energy. This fact supports the validity of the two-step model for high-energy photons.

On the other hand, the two-step model breaks down for the low-energy region. As can be seen in Fig. 4, the L-electron shakeoff probability increases more rapidly with energy than the K-shell shakeoff probability. The breakdown of the two-step model occurs at approximately 10 for the L₁-shell electron and at about 5 for the L₂-shell case. However, in the case of the K-shell shakeoff the probability increases more gradually and the energy dependence of the probability is more important.

Sachenko and Burtsev¹⁸⁾ have made nonrelativistic calculations on the energy dependence of the L-shell shakeoff probability during K-shell photoelectric effect. They used the screened hydrogenic wave functions with the Slater's screening constants and calculations were performed for Al, K, and Fe. Their results indicate that the shakeoff probability approaches to the constant value at the reduced energy of 1.2-1.3. This energy is about 4 times smaller than the breakdown energy obtained above.

In the present work, we neglect the effect of the electron exchange, as described above. This assumption is adequate for high-energy photons. However, in the low-energy region close to the threshold energy of the shakeoff process the energy of the photoelectron is also low and near to that of the shakeoff electron. For such a case, the exchange effect seems to be important. We have shown in the case of the K-shell shakeoff process in nuclear β decay that inclusion of the exchange effect enhances the shakeoff probability in the low-energy region.¹⁹⁾ If we take into account the effect of electron exchange, the shakeoff probability for low-energy photons may increase and the breakdown energy of the two-step model may become lower.

Recently the experimental studies on the K-shell excitation probabilities accompanying K-shell photoionization have been carried out by Keski-Rahkonen et al.²⁰⁾ They irradiated the targets of Mg, V, Cr, Mn, and Fe by x rays and observed the intensity of the K₂^h hypersatellite lines, which originate from the double K-shell vacancies, by the use of a plane crystal Bragg spectrometer. The experimental values they obtained are actually not the K-shakeoff probabilities, but the double K-hole creation probabilities, which include the contribution from the shakeup process.

Table 1. Comparison of the calculated and measured K-electron shakeoff probabilities accompanying K-shell photoionization ($\times 10^{-4}$).

Element	Experimental ^{a)}	Theoretical			
		Present	Two-step ^{b)}	SA ^{c)}	A ^{d)}
Mg	14	1.65	4.77	7.2	18
V	3.1	0.86	1.84	1.9	4.6
Cr	9.2	0.73	1.71	1.7	4.2
Mn	9.7	0.61	1.60	1.6	3.9
Fe	5.5	0.50	1.50	1.5	3.6

a) Keski-Rahkonen et al. (Ref. 20).

b) Two-step calculation in the present model.

c) Suvanen and Åberg (Ref. 21).

d) Åberg (Ref. 22), including the shakeup probabilities.

In Table 1, the experimental results of Keski-Rahkonen et al. are compared with the present theoretical calculations. The photon energies used for the one-step model is 5.4 keV (Cr $K\alpha$) for Mg and 30 keV for other elements.²⁰ The values obtained from the two-step calculations are also included in the table. It is clear that the energy dependence of the shakeoff probability is appreciable and the one-step values are less than one half of the two-step ones. For comparison, the theoretical values calculated by Åberg and Suvanen²¹ and by Åberg²² are listed in the table. These theories are nonrelativistic and based on the two-step model. Åberg and Suvanen²¹ used the correlated wave functions for helium-like ions in the generalized sudden approximation [Eq. (7) of the paper of Åberg²³]. Their values in the table are obtained using a 9-parameter wave functions of Hylleraas.²⁴ On the other hand, the calculations of Åberg²² are also made using the correlated wave functions, but include the contributions from the shakeup process.

The two-step results in the present work are in good agreement with the values of Åberg and Suvanen,²¹ except for Mg. It is interesting to note that the present relativistic theory neglect the effect of electron correlation, while the model of Åberg and Suvanen includes the correlation effect but nonrelativistic.

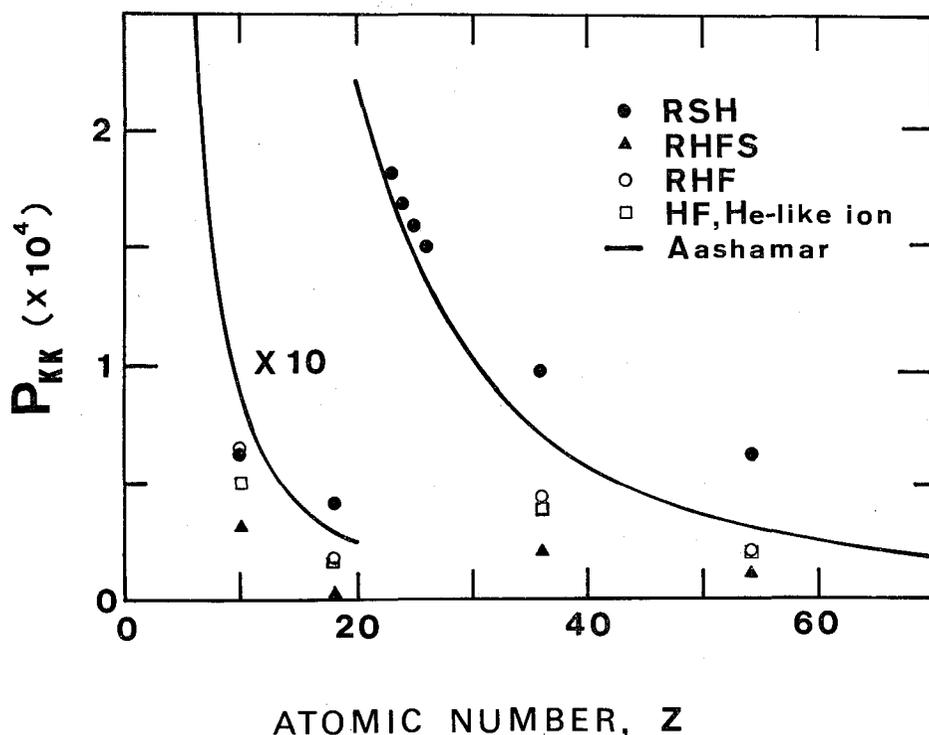


Fig. 5. K-shell shakeoff probability as a function of atomic number. The solid circles represent the present relativistic screened hydrogenic (RSH) results in the two-step approach. The solid triangles indicate the RHFS calculations of Carlson and Nestor (Ref. 1). The open circles and squares correspond to the RHF results and the HF results for helium-like ions (Ref. 25). The solid curve gives the results obtained with the 204-parameter Hylleraas-type wave functions of Aashamar (Ref. 25).

They also calculated the total excitation probability including the ionization and excitation processes, but the obtained results are almost equal to the values of Åberg cited in Table 1. Better agreement of the total excitation cross sections with the experimental data may suggest that there are large contributions from the shakeup process. However, further more experimental studies are needed to estimate the contribution from the shakeup process in double K-vacancy production.

Recently Pletonen and Åberg²⁵⁾ have calculated the K-shell excitation and ionization probabilities in K-electron capture decay and in K-shell internal conversion as a function of atomic number Z in the generalized sudden approximation.²⁵⁾ They used a 204-parameter Hylleraas-type wave function²⁶⁾ of helium-like ions for the initial state and the hydrogenic wave function for the final excited state. Comparison of these results with the nonrelativistic HF and the relativistic HF (RHF) calculations reveals that the effect of electron correlation is more important than the relativistic effect. Since their model is based on the two-step model, the K-shell ionization probability in K-shell conversion can be interpreted as that in K-shell photoionization.

In Fig. 5, the K-shell shakeoff probabilities in the two-step model are compared with various calculations of Pletonen and Åberg.²⁵⁾ The solid circles represent the relativistic screened hydrogenic calculations (RSH) of the present work, while the solid triangles correspond to the results of Carlson and Nestor.¹⁾ The open circles and squares indicate the RHF results²⁵⁾ and the HF calculations for helium-like ions,²⁵⁾ respectively. The solid line gives the results obtained using the 204-parameter Hylleraas-type wave functions of Aashamar.²⁵⁾ The present results are in good agreement with the values calculated using the Hylleraas-type wave function for low- and intermediate- Z elements. However, in the case of Xe the present model yields higher value than the value of Pletonen and Åberg.

ACKNOWLEDGMENT

The author would like to express his thanks to Prof. T. Åberg for sending his unpublished results.

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