Calculation of K-shell ionization cross sections by charged particles in the distortion approximation using pseudostates

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Cross sections for K-shell ionization by charged particles have been calculated within the framework of the impact-parameter formalism using the distortion approximation. The continuum wave functions are represented by pseudostates. It is confirmed that the pseudostate wave function can well reproduce the shape of the continuum wave function for low-energy electrons. The calculated total K-shell ionization cross sections for a number of ion-atom collision systems are shown to be in good agreement with the experimental results. For protons on copper, the ionization probabilities as a function of impact parameter are also found to agree with the measured values. The present results indicate that the distortion approximation using pseudostates is very useful in the study of K-shell ionization cross sections over a large range of ion-atom collision systems.

I. INTRODUCTION

Inner-shell ionization by charged-particle impact has been extensively studied in recent years.[1] Various simple perturbative models, such as the plane-wave Born approximation[2] (PWBA) and the semiclassical approximation[3] (SCA), have been used to calculate the ionization cross sections. It is well known that corrections to PWBA and SCA, such as the Coulomb-deflection effect, the binding-energy effect, the electronic relativistic effect, and the polarization effect should be taken into account to interpret the experimental data.

In the calculation of ionization cross sections, the final states are represented by continuum wave functions. However, calculations of transition matrix elements involving continuum wave functions are quite tedious. Even in the simplest hydrogenic potential, the continuum wave functions are expressed in terms of confluent hypergeometric functions. Application of the ionization theory to higher-order approximations than the first-order Born theory is generally limited by this difficulty.

From a computational point of view, it is often more convenient to replace the continuum by a set of discrete wave functions, called *pseudostates*. These states are obtained by diagonalizing a Hamiltonian in a set of discrete basis functions. Pseudostates are the states with positive eigenvalues. This technique can be used with any finite sets of any square-integrable (L^2) basis functions.

A main advantage of the pseudostate method is its simplicity because all the wave functions, both for bound and free states, are expressed in terms of L^2 basis functions. It is especially advantageous when we have to go beyond the first-order Born approximation, such as the distortion approximation and the coupled-state calculations. Another advantage lies in the fact that this method can be readily applied not only for the hydrogenic potential, but also to arbitrary atomic potentials.

In recent years, the pseudostate method has been used successfully for inner-shell ionization processes, such as K-hole production cross sections by light ions,[4] double-center expansion method in proton-hydrogen collisions,[5,6] He^{2+} on Li,[7] and energy distribution of ejected electrons in ion-atom collisions.[8]

The purpose of the present work is to examine the validity of the simple distortion approximation[9] in the calculation of K-shell ionization cross sections in ion-atom collisions by the use of the pseudostate method. The computed results are compared with the experimental data as well as with other theoretical calculations.

II. METHOD

We consider the system in which the target atom is located at the fixed origin and the projectile is moving along a classical trajectory with a constant speed. The Schrödinger equation for the system is

$$\left[H-i\frac{\partial}{\partial t}\right]\Psi(\mathbf{r},t)=0 , \tag{1}$$

where $\mathbf{r}$ is the coordinate of the target electron and the Hamiltonian is given by

$$H=H_0+V(\mathbf{r},\mathbf{R}) . \tag{2}$$

Here the unperturbed Hamiltonian of the target atom H_0 and the time-dependent perturbation $V(\mathbf{r},\mathbf{R})$ are

$$H_0=-\tfrac{1}{2}\nabla^2+U(r) , \tag{3}$$

$$V(\mathbf{r},\mathbf{R})=-\frac{Z_1}{|\mathbf{R}-\mathbf{r}|} , \tag{4}$$

where $U(r)$ is the atomic potential, $\mathbf{R}(t)$ is the prescribed trajectory of the projectile, and Z_1 is the atomic number of the projectile. Atomic units ($e=m_e=\hbar=1$) are used

throughout the present work.

According to the distortion approximation,[9] the transition probability of the target electron from an initial state 0 to a final state n by the projectile with impact parameter b is given by

$$P_n(b)=\left|\int_{-\infty}^{\infty} V_{n0}(t)\exp[-i\eta(t)]dt\right|^2 , \quad (5)$$

where the distortion phase is defined as

$$\eta(t)=\int_{-\infty}^{t}\{[\varepsilon_n+V_{nn}(t')]-[\varepsilon_0+V_{00}(t')]\}dt' . \quad (6)$$

The matrix element V_{nk} is expressed as

$$V_{nk}=-\int d\mathbf{r}\,\phi_n^*(\mathbf{r})V(\mathbf{r},\mathbf{R})\phi_k(\mathbf{r}) , \quad (7)$$

where ε_k and ϕ_k represent the energy eigenvalue and eigenfunction of the state k for the single-particle Hamiltonian H_0:

$$H_0\phi_k=\varepsilon_k\phi_k . \quad (8)$$

The total ionization cross section is given by

$$\sigma=2\pi\int_0^{\infty}P_n(b)b\,db . \quad (9)$$

We expand the eigenfunction $\phi_k(\mathbf{r})$ in terms of a set of L^2 basis functions $\chi(r)$,

$$\phi_{jlm}(\mathbf{r})=\sum_{i=1}^{N} c_i\chi_i(r)Y_{lm}(\hat{\mathbf{r}}) , \quad (10)$$

where l and m are the orbital and magnetic quantum numbers, $Y_{lm}(\hat{\mathbf{r}})$ is the spherical harmonics, and $\hat{\mathbf{r}}$ is the unit vector in the direction of $\mathbf{r}$. We choose Slater-type orbitals (STO's) as basis functions:

$$\chi_i(r)=N_i r^{n_i-1}\exp(-\zeta_i r) , \quad (11)$$

with

$$N_i=\frac{(2\zeta_i)^{n_i+1/2}}{\Gamma(2n_i+1)^{1/2}} .$$

Using a finite set of these basis functions, the Hamiltonian H_0 is diagonalized to obtain the energy eigenvalues ε_k and eigenfunctions ϕ_k. The atomic potential $U(r)$ in Eq. (3) is calculated by the Hartree-Fock-Slater (HFS) method,[10] except in the case of the hydrogen atom. With a suitable choice of the number of STO's in the basis set, N, the principal quantum numbers, n_i, and orbital exponents, ζ_i, of the STO's, we can reproduce the HFS energy eigenvalues with 0.1% accuracy.

In addition to the bound atomic states, there exist positive discrete energy eigenvalues of H_0. These pseudostates are used to calculate inner-shell ionization cross sections. The total ionization cross section is the sum of the cross sections for excitation to these discrete pseudostates. As pointed out earlier,[6] the precise form of the STO's is not important, but we ensure that there are at least a few pseudostates within 1–2 a.u. above the ionization threshold.

III. RESULTS AND DISCUSSION

To test the quality of pseudostates, we compare these discrete functions with the correct continuum wave functions with the same energy eigenvalues. We note that Yamani and Reinhardt[11] have shown that for certain limited cases the pseudostate wave function is proportional to the continuum wave function.

Comparison between the pseudostate and continuum wave functions for various l values and energies E is made for the hydrogen atom, where the correct form of the continuum wave function is well known:[12]

$$\psi_E=\frac{N_{kl}}{kr}\frac{2^l e^{-\pi\eta/2}|\Gamma(l+1+i\eta)|}{\Gamma(2l+2)}(kr)^{l+1}e^{-ikr} \times F(l+1-i\eta,2l+2;2ikr) . \quad (12)$$

Here k is the wave number, $\eta=-Z/k$, and $F(a,b;z)$ the confluent hypergeometric function. The basis set used consists of ten STO's for $l=0$ and seven STO's for $l=1$, respectively. The parameters of the STO's and the energy eigenvalues obtained by diagonalizing the Hamiltonian are listed in Table I.

Figure 1 shows the comparison of pseudostate and continuum wave functions for $l=0$ and 1. The pseudostate wave functions are normalized to the continuum ones at the first peak. It is clear that in the inner region and for low energies each pseudostate wave function reproduces the shape of the continuum wave function. For higher energies, the latter oscillates more rapidly within the inner region and this behavior can be better represented if the size of the STO basis set is enlarged. However, the main contribution to the inner-shell ionization cross section comes from the low-energy region and the discrepancy at high energies is not very important.

Using the STO basis set similar to Table I, we have calculated the K-shell ionization cross sections for protons on hydrogen atom in the distortion approximation. For comparison, we have also made calculations in the first Born approximation, which is obtained from Eq. (6) if $\eta(t)$ is replaced by $(\varepsilon_n-\varepsilon_0)t$. This corresponds to the SCA by the use of pseudostates. All the calculations include partial-wave contributions from $l=0$ to 2. The cal-

TABLE I. Principal quantum numbers n and orbital exponents ζ for the Slater-type basis functions and the energy eigenvalues E (a.u.) obtained by diagonalizing the hydrogen Hamiltonian.

	$l=0$			$l=1$	
n	ζ	E	n	ζ	E
1	1.0	−0.5	2	0.5	−0.125
1	0.5	−0.125	2	0.3333	−0.0555
2	0.5	−0.0555	3	0.3333	−0.0242
3	0.5	−0.0203	2	0.8	0.0657
4	0.5	0.0483	2	1.125	0.3845
1	0.8	0.2054	2	1.5625	1.4920
1	1.25	0.5684	2	2.4414	6.8227
1	1.4531	1.4866			
1	1.5625	4.4092			
1	2.4414	21.6011			

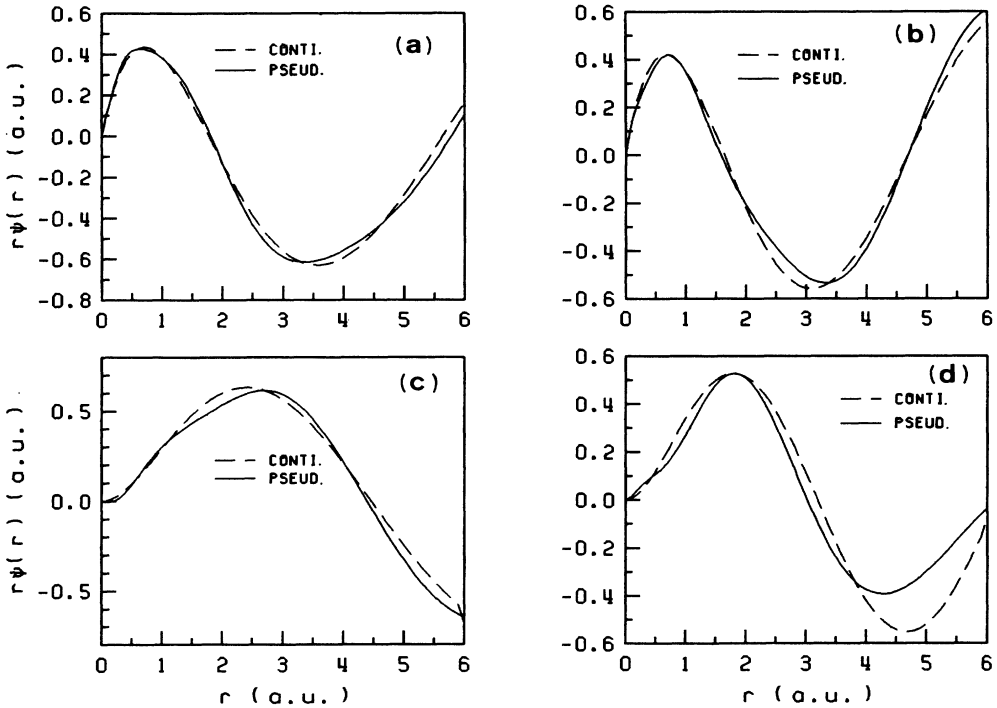

FIG. 1. Comparison of pseudostate wave functions of hydrogen atom with Coulomb continuum wave functions. The solid curve represents the pseudostate wave function and the dashed curve indicates the correct continuum one. (a) $l=0$ and $E=0.045$ a.u., (b) $l=0$ and $E=0.205$, (c) $l=1$ and $E=0.066$, and (d) $l=1$ and $E=0.384$.

culated results are shown in Fig. 2 and compared with experimental data and other theoretical models.

It can be seen from the figure that the present result in the distortion approximation agrees with the experimental values for energy region above 40 keV (Fite *et al.*,[15] Park *et al.*,[16] and Shah and Gilbody[17]). At lower energies the present values are slightly larger than the more recent experimental data of Shah, Elliot, and Gilbody.[18] In such a low-energy region, the effect of charge transfer channels becomes important and more elaborate theoretical treatments are necessary. On the other hand, as is well known, the first Born approximation agrees well with experiments at higher energies, but yields a gross overestimate in the low-energy region.

In the figure, other theoretical calculations which include the effect of charge transfer channels are shown. The method based on the time-dependent Schrödinger equation (TDSE) by Terlecki, Grün, and Scheid[13] gives larger values than the experimental data. Fritsch and Lin[6] made coupled-state calculations based on a two-center expansion in atomic orbitals and pseudostates. Their results with 46 atomic states plus pseudostates are closer to the experimental data of Shah, Elliot, and Gilbody[18] in the low-energy region than the present values, but are smaller than the experiment at high energies. The main reason for the discrepancy in the high-energy region is ascribed to the fact that their results contain only the contributions from $l=0$ and 1. The two-center calculations of Shakeshaft,[5] using a 70-scaled-hydrogenic-state basis set up to $l=2$, are in good agreement with the present results in the energy region higher than 40 keV. At lower energies, the trend of his results is similar to that of Fritsch and Lin.[6] Winter and Lin[14] carried out the calculations with a triple-center atomic state method for the energy region below 15 keV. This latter calculation includes effects due to the saddle point of the

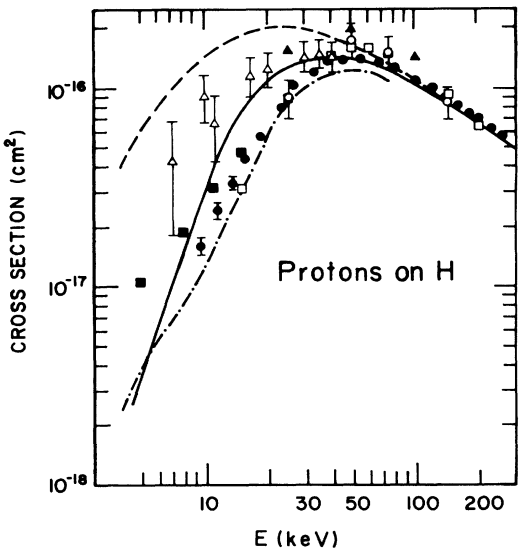

FIG. 2. Ionization cross sections of hydrogen atom by protons. Theory: ——, distortion approximation (present work); — — —, Born approximation (present work); —·—·—·, two center (Ref. 6); □, two center (Ref. 5); ■, triple center (Ref. 14); ▲, TDSE (Ref. 13). Experiment: △ (Ref. 15); ○ (Ref. 16); ● (Refs. 17 and 18).

potential surface which is considered to be important in low-energy collisions. Their results are somewhat higher than experimental data below 10 keV.

Figure 3 shows the comparison of theoretical and experimental ionization cross sections for He^{2+} on He. Within the independent-electron approximation, the HFS potential for the helium atom was used to obtain the initial-state and pseudostate wave functions. Contributions from the continuum electrons with angular momentum up to $l=2$ were considered. The basis set consists of ten STO's for $l=0$, ten STO's for $l=1$, and ten STO's for $l=2$. The present results are in good agreement with the recent experimental values of Shah and Gilbody.[21]

For this system, Sidorovich and Nilolaev[19] used the Born ionization probabilities for hydrogenlike atoms with effective nuclear charges and calculated the total ionization cross sections. Their results are indicated by the dot-dashed curve in Fig. 3. They also unitarized the ionization probabilities according to the decay model and found that the unitarization decreases the ionization cross sections. The unitarized Born cross sections are shown in the figure by the dashed curve. Both models are in agreement with the present values and experimental data in the high-energy region, but at low energies their results are substantially larger.

A similar comparison for Li^{3+}-ion impact on He is shown in Fig. 4. It can be seen that the distortion approximation agrees well with the experimental results, while both Born and unitarized Born cross sections by Sidorovich and Nikolaev[19] overpredict the experimental data in the low-energy region. From Figs. 2–4, it can be said that the distortion approximation with pseudostates can give the K-shell ionization cross sections in agreement with experiments for symmetric and nearly symmetric collisions.

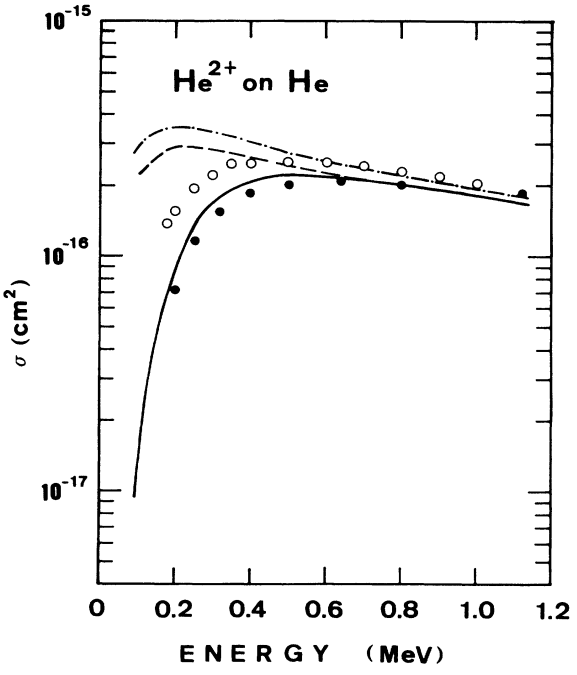

FIG. 3. Ionization cross sections of He by He^{2+}. Theory: ——, distortion approximation (present work); —·—·—·, Born (Ref. 19); — — —, unitarized Born (Ref. 19). Experiment: ○ (Ref. 20); ● (Ref. 21).

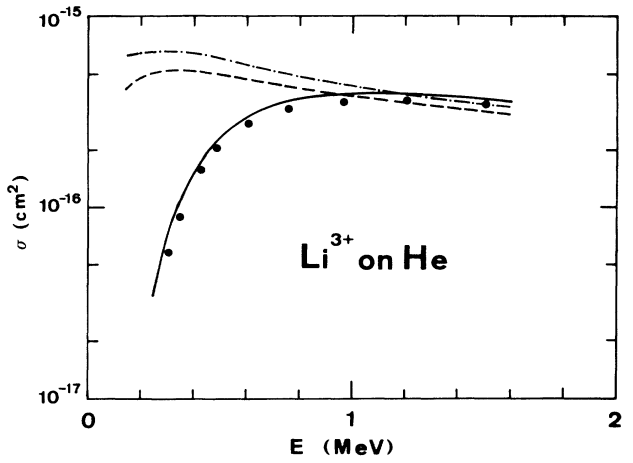

FIG. 4. Ionization cross sections of He by Li^{3+}. Theory: ——, distortion approximation (present work); —·—·—·, Born (Ref. 19); — — —, unitarized Born (Ref. 19). Experiment: ● (Ref. 21).

We have also applied the present model for the K-shell ionization cross sections for asymmetric collisions. The calculations were made for the case of proton impact on copper. The atomic potential was obtained in the HFS approximation. The number of STO's for each l was chosen to be ten for $l=0$, 1, and 2.

In Fig. 5 the calculated K-shell ionization probabilities for 0.5-MeV protons on copper are shown as a function of impact parameters. The solid curve is from the distortion approximation, and the dotted curve is from the

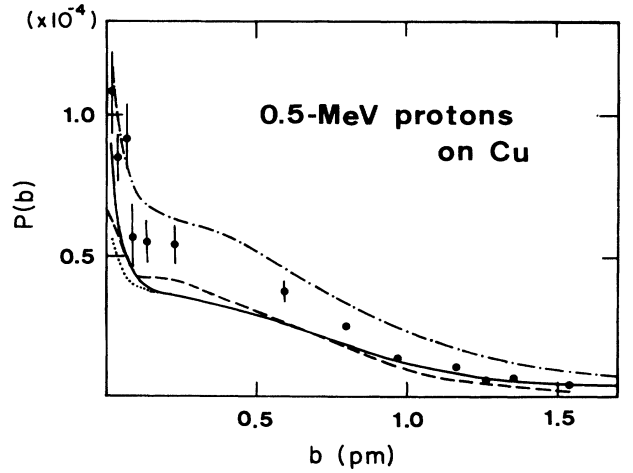

FIG. 5. Ionization probabilities as a function of impact parameter for 0.5-MeV protons on Cu. Theory: ——, distortion approximation (present work); · · · ·, distortion approximation with recoil effect (present work); —·—·—·, Born approximation (present work); — — —, coupled-state calculations (Ref. 23). Experimental: ● (Ref. 24).

same approximation but with the inclusion of recoil effect, which was treated in the manner similar to Amundsen.[22] The dot-dashed curve is from the Born approximation, which is similar to the SCA except for the use of pseudostates. All the calculations were performed assuming the Rutherford trajectory for the projectile. The dashed curve is from the coupled-state calculations of Reading *et al.*[23] with the inclusion of the recoil effect. The experimental data are taken from Andersen *et al.*[24]

It is clear that the distortion approximation is in good agreement with the coupled-state calculation, while the Born approximation yields larger values than these two models. The experimental results are in agreement with the distortion approximation at large impact parameters, but are about 20% higher for small impact parameters.

The present results of total K-shell ionization cross sections for 0.5-, 1.0-, and 2.0-MeV protons on copper are compared in Table II with other theoretical results as well as with the experimental data and the recommended reference cross sections of Paul and Sacher.[29] The PWBA cross sections are obtained according to Merzbacher and Lewis[2] and the PWBA values with corrections for the binding-energy and Coulomb-deflection effects (PWBA-BC) are evaluated by the method of Brandt and Lapicki.[25] Both cross sections were calculated by the use of the DEKY code[30] and the energy-loss effect is correctly taken into account. However, the electronic relativistic effect is neglected because the present results are nonrelativistic.

The present Born results are in good agreement with the PWBA values, while the distortion approximation agrees well with the PWBA-BC. The latter result means that the distortion approximation accounts for all the binding-energy correction which was included semiempirically in the PWBA-BC. The values in the distortion approximation are in agreement with the experimental values of Lear and Gray,[26] but smaller than the more recent experimental data of Laegsgaard, Andersen, and Høgedal[27] and Sera *et al.*,[28] and also the reference data of Paul and Sacher.[29] This discrepancy can be attributed to the electronic relativistic effect,[31] which is neglected in the present work.

TABLE II. Comparison of K-shell ionization cross sections for protons on Cu (in units of 10^{-24} cm^2).

Source	Energy (MeV) 0.5	1.0	2.0
Born[a]	2.93	22.3	111
PWBA[b]	2.88	22.3	113
DA[c]	1.44	14.9	90.1
PWBA-BC[d]	1.41	14.3	85.8
Experiment	1.134±0.227[e]	14.545±0.909[e]	89.1±6.6[e]
	1.72±1.7[f]	16.0±0.5[f]	96.2±2.9[f]
	1.58±0.19[g]		
Reference[h]	1.618	15.84	99.14

[a]Born approximation with straight-line trajectory, present result.
[b]Plane-wave Born approximation, Ref. 2.
[c]Distortion approximation with Coulomb trajectory, present result.
[d]PWBA with binding-energy and Coulomb-deflection corrections, Ref. 25.
[e]Lear and Gray, Ref. 26.
[f]Laegsgaard, Andersen, and Høgedal, Ref. 27.
[g]Sera *et al.*, Ref. 28.
[h]Reference data, Ref. 29.

IV. CONCLUSION

We have calculated the K-shell ionization cross sections in ion-atom collisions by the use of pseudostates in the distortion approximation. It is found that the pseudostate wave functions can well reproduce the behavior of the correct Coulomb continuum wave functions in the low-energy region and for small radial distances. The use of pseudostates to represent continuum wave functions makes it possible to extend the distortion approximation to inner and outer-shell ionizations from nonhydrogenic targets. In the present paper the initial- and final-state wave functions are calculated from the Hartree-Fock-Slater potential of the target atom.

We have applied this method to calculate single ionization cross sections for proton impact on hydrogen atoms, and for He^{2+} and Li^{3+} on He within the independent-electron model. Our results indicate that the distortion approximation gives correct ionization cross sections over a large energy range. It is interesting to note that the predicted cross sections are in good agreement with experimental results even in the energy region below where the cross section peaks. This agreement is somewhat surprising since it is known that in this low- and intermediate-energy region, charge transfer channels are dominant, with the ionizations being the small channels. It is not understood why the perturbative calculation within the distortion approximation can give good ionization cross sections without including the effects of charge transfer channels, such as those treated in the coupled-channel methods.

We have also applied the distortion approximation to calculate K-shell ionization probabilities and cross sections for protons on copper. One expects that for such asymmetric collision systems a perturbative approach, such as the distortion approximation, will be valid. The calculated ionization probabilities are in good agreement with experimental results at large impact parameters, but discrepancies still exist at small impact parameters whether the recoil effects are included or not. However, the present results agree well with the results of coupled-channel calculations over the whole range. In the case of total K-shell ionization cross sections, the calculated results are about 10% smaller than the reference cross sections and the recent experimental data. This discrepancy is due to the electronic relativistic effects. We have already shown that it is possible to obtain the relativistic pseudostates for realistic atomic potentials.[32] The calculations of K-shell ionization cross sections in ion-atom collisions using these relativistic pseudostates are in progress.

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