Electron-impact ionization of uranium atomic ions

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Electron-impact ionization cross sections for U^{16+} and U^{82+} are calculated in the distorted-wave approximation. Excitation-autoionization contributions are found to enhance the direct ionization cross section for U^{16+} by a factor of 3 in the threshold energy region. Excellent agreement is found between the calculated total ionization cross section for U^{16+} and recent crossed-beams experimental measurements. Only the direct ionization cross section for U^{82+} is calculated since excitation-autoionization contributions are assumed to be small due to strong radiation field damping.

I. INTRODUCTION

Distorted-wave theory has proved quite successful in treating the electron-impact ionization of atomic ions.[1] For incident energies near the binding-energy threshold, the direct ionization cross section may be calculated using a triple partial-wave expansion of the first Born scattering amplitude.[2-6] The distorted-wave approximation may then be used to calculate the many incident, ejected, and scattered partial waves. In the threshold energy region, indirect ionization by excitation-autoionization will sometimes make substantial contributions to the total ionization cross section.[7-12] The distorted-wave approximation may again be used to calculate the partial waves needed to evaluate the excitation cross section to autoionizing levels.

In recent years relativistic distorted-wave theory has been employed to calculate ionization processes in heavy atomic ions.[13,14] In this paper we examine electron-impact ionization for atomic ions in the uranium isonuclear sequence. In Sec. II relativistic distorted-wave theory as applied to electron ionization of atomic ions is briefly reviewed. In Sec. III we present cross section results for both U^{16+} and U^{82+}, and then further compare the U^{16+} cross section with experiment. A brief summary is contained in Sec. IV.

II. THEORY

The general form of the direct ionization cross section (in atomic units) may be written as[14]

$$\sigma_{\rm dir}=\int_0^{E/2}\frac{16W}{p_i^3 p_e p_f}\sum_{l_i}\sum_{l_e}\sum_{l_f}\left[\sum_{\lambda}\Omega_d^{\lambda}+\sum_{\lambda'}\Omega_e^{\lambda'}-\sum_{\lambda}\sum_{\lambda'}|\Omega_{\rm int}^{\lambda\lambda'}|\right]d\varepsilon_e\ , \quad (1)$$

where the partial-wave expansion is over the incident, ejected, and final scattered angular momenta (l_i,l_e,l_f). The direct, exchange, and interference product matrix elements $(\Omega_d,\Omega_e,\Omega_{\rm int})$ are evaluated in a multipolar expansion (λ). The maximum ejected energy is E and the maximum-interference approximation of Peterkop[2] is imposed by taking the negative of the absolute value of the third term in Eq. (1).

For the evaluation of nonrelativistic and semirelativistic direct ionization cross sections, the product matrix elements Ω in Eq. (1) are evaluated in a configuration-average approximation.[14] The occupation number of the nl subshell to be ionized in the initial configuration is W, the energy-momentum relation is $p=\sqrt{2\varepsilon}$, and the continuum normalization is one times a sine function. For nonrelativistic cross sections the energies and radial orbitals needed to evaluate the matrix elements are calculated in the Hartree-Fock (HF) approximation.[15] For semirelativistic cross sections the energies and radial orbitals needed to evaluate the matrix elements are calculated in the Hartree-Fock approximation with relativistic modifications (HFR),[15] which includes the mass-velocity and Darwin corrections within modified differential equations.[16]

For the evaluation of fully relativistic direct ionization cross sections, the product matrix elements Ω in Eq. (1) are evaluated in a subconfiguration average approximation.[14] The number of terms in each partial-wave expansion doubles since for each l there are $j=l\pm\frac{1}{2}$ components. The occupation number of the nlj subshell to be ionized in the initial configuration is W, the energy-momentum relation is $p=(2\varepsilon+\alpha^2\varepsilon^2)^{1/2}$, and the continuum normalization is $(1+\alpha^2\varepsilon/2)^{1/2}$ times a sine function $(\alpha=\frac{1}{137})$. The energies and radial orbitals needed to evaluate the matrix elements are calculated in the Dirac-Fock (DF) approximation.[17] Each radial matrix element contains contributions form both components of the Dirac bispinor.

The general form of the excitation cross section needed to evaluate indirect ionization contributions may be written as[11]

$$\sigma_{\rm exc}=8\pi\frac{W_1(W_2^f-W_2)}{p_i^3 p_f}\times\sum_{l_i}\sum_{l_f}\left[\sum_{\lambda}\Omega_d^{\lambda}+\sum_{\lambda'}\Omega_e^{\lambda'}-\sum_{\lambda}\sum_{\lambda'}\Omega_{\rm int}^{\lambda\lambda'}\right]\ , \quad (2)$$

where the partial-wave expansion is over the incident and final scattered angular momenta (l_i,l_f).

For the evaluation of nonrelativistic and semirelativistic excitation cross sections, the product matrix elements Ω in Eq. (2) are evaluated in a configuration-average ap-

proximation.[11] For the transition $nl \rightarrow n'l'$, W_1 is the occupation number of the nl subshell in the initial configuration and W_2 is the occupation number of the $n'l'$ subshell in the initial configuration ($W_2^f = 4l'+2$). The energy-momentum relation is $p = \sqrt{2\varepsilon}$ and the continuum normalization is one times a sine function. The energies and radial orbitals are calculated in the same manner as for the nonrelativistic or semirelativistic direct ionization cross section.

For the evaluation of fully relativistic excitation cross sections, the product matrix elements Ω in Eq. (2) are evaluated in a subconfiguration-average approximation. For the transition $nlj \rightarrow n'l'j'$, W_1 is the occupation number of the nlj subshell in the initial subconfiguration and W_2 is the occupation number of the $n'l'j'$ subshell in the initial subconfiguration ($W_2^f = 2j'+1$). The energy-momentum relation is $p = (2\varepsilon + \alpha^2\varepsilon^2)^{1/2}$, and the continuum normalization is $(1+\alpha^2\varepsilon/2)^{1/2}$ times a sine function. The energies and radial orbitals are calculated in the same manner as for the fully relativistic direct ionization cross section.

III. CROSS-SECTION RESULTS

Direct ionization contributions to the total ionization of the $5d^8$ ground configuration of U^{16+} were calculated using Eq. (1) in both the HF and HFR approximations for the four outer subshells: $5d$, $5p$, $5s$, and $4f$. The remaining inner subshells will contribute to multiple ionization. Cross-section results are presented in Table I at twice the threshold energy. For the $5d$ and $4f$ subshells, relativistic effects lower the ionization potential and raise the cross section, while for the $5p$ and $5s$ subshells just the opposite result occurs. The largest relativistic effects occur for the $5s$ subshell; the ionization potential is raised by 76.5 eV and the cross section is lowered by 20%. Cross-section changes in the other subshells are less than 10%.

Excitation-autoionization contributions to the total ionization of the $5d^8$ ground configuration of U^{16+} were calculated using Eq. (2) in the HFR approximation for 69 excitations: $5p \rightarrow 7f$, $5p \rightarrow n'l'$ ($n'=8,9,10$; $l'=0,1,2,3$), $5s \rightarrow 6l'$ ($l'=2,3$), $5s \rightarrow n'l'$ ($n'=7,8,9,10$; $l'=0,1,2,3$), $4f \rightarrow 5l'$ ($l'=3,4$), $4f \rightarrow n'l'$ ($n'=6,7,8,9,10$; $l'=0,1,2,3,4,5$), $4d \rightarrow 5l'$ ($l'=2,3$), and $4d \rightarrow 6l'$ ($l'=0,1,2,3$). An n^3 rule extrapolation method was used to estimate contributions from the excitations $5p \rightarrow n'l'$, $5s \rightarrow n'l'$, and $4f \rightarrow n'l'$ for $n' > 10$. Cross-section results for the four largest contributions are presented in Table II at the threshold energy in both the HF and HFR approximations. For the four excitations presented, relativistic effects lower the threshold energy and raise the cross section. For the $4f \rightarrow 5f$ excitation the threshold energy is lowered by 27.3 eV and the cross section is raised by 9%.

The total single ionization cross section, ignoring interference effects between the direct ionization and excitation-autoionization processes, may be written as

$$\sigma_{\text{tot}}(i) = \sum_f \sigma_{\text{ion}}(i \rightarrow f) + \sum_j \sigma_{\text{exc}}(i \rightarrow j) B_j^a , \tag{3}$$

where $\sigma_{\text{ion}}(i \rightarrow f)$ is the direct ionization cross section from the initial configuration to a particular level f within the ionized configurations, $\sigma_{\text{exc}}(i \rightarrow j)$ is the excitation cross section from the initial configuration to a particular level j within the core-excited configurations, and B_j^a is the branching ratio for autoionization from level j. The further indirect ionization process of resonant-recombination double autoionization is not included.

The total ionization cross section from the $5d^8$ ground configuration of U^{16+} was calculated using Eq. (3) in the HFR approximation. The configuration-average cross sections calculated using Eqs. (1) and (2), and discussed in the first two paragraphs of this section, were either statistically partitioned over all levels of the final ionized or core-excited configurations and then summed taking explicit account of the energy position of each ionized or core-excited level calculated using the atomic structure package of Cowan,[15] or, in the case of relatively small cross sections, summed at the configuration-average energy. For the strong $4f \rightarrow 5f$ excitation, the $4f^9 5s^2 5p^6 5d^8 5f$ core-excited configuration contains 954 levels distributed over an energy range of 70.3 eV. Even though U^{16+} has a relatively high net charge, we assumed that $B_j^a = 1$ for each level due to the large number of autoionizing decay channels present in core-excited states formed from the $5d^8$ ground configuration. The

TABLE I. Direct ionization cross sections for U^{16+}.

Subshell	Ionization potential (eV)	Cross section at twice threshold (10^{-18} cm^2)
	Hartree-Fock	
$5d$	383.99	0.859
$5p$	483.91	0.347
$5s$	541.13	0.084
$4f$	723.25	0.665
	Hartree-Fock relativistic	
$5d$	381.27	0.882
$5p$	508.16	0.324
$5s$	617.63	0.067
$4f$	689.67	0.714

TABLE II. Excitation cross sections to autoionizing configurations for U^{16+}.

Excitation	Threshold energy (eV)	Threshold cross section (10^{-18} cm^2)
	Hartree-Fock	
$4f \rightarrow 5f$	448.57	0.882
$4f \rightarrow 5g$	549.43	0.284
$4f \rightarrow 6f$	559.30	0.182
$4f \rightarrow 6g$	602.06	0.178
	Hartree-Fock relativistic	
$4f \rightarrow 5f$	421.24	0.962
$4f \rightarrow 5g$	517.70	0.294
$4f \rightarrow 6f$	527.48	0.204
$4f \rightarrow 6g$	569.66	0.190

assumption is based on detailed radiative and autoionization rate calculations for open-shell ions in the Fe isonuclear sequence.[18]

In Fig. 1 the HFR total ionization cross section for U^{16+} is compared with the experimental crossed-beams measurements of Gregory *et al.*[19] The overall agreement is excellent. In the energy region from 400 to 500 eV experimental evidence may be seen for possible resonant-recombination double autoionization (RRDA) processes. Work remains in identifying the energy position of possible configurations formed by attachment of an incident electron to a core-excited target state. Even if clear identifications can be made, however, the challenge of carrying out a calculation for the magnitude of the RRDA process in such a complicated atomic structure case remains formidable.

Direct ionization contributions to the total ionization of the $2p^6$ ground configuration of U^{82+} were calculated using Eq. (1) in the HF, HFR, and DF approximations for both the $2p$ and $2s$ subshells. Cross-section results are presented in Table III at twice the threshold energy. The HFR calculations are found to increase the $2p$ and $2s$ ionization potentials over the HF results by 6% and 18%, respectively. When the $2p(3/2)$ and $2p(1/2)$ ionization potentials are averaged, there is good agreement between the HFR and DF calculations for the ionization potential. The HFR calculations give a 25% reduction in the HF results for the $2p$ cross section, and a 42% reduction in the $2s$ cross section. However, when the $2p(3/2)$ and $2p(1/2)$ cross sections are summed, the DF calculations are about the same as the HF results for the $2p$ cross section, and give only a 22% reduction in the $2s$ cross section. As previously discussed,[14] the HFR method for ionization cross sections, as formulated in Sec. II, neglects the effect of the small component of the Dirac bispinor on both continuum normalization and the calculation of radial matrix elements. As these results show for L-shell ionization from heavy atomic systems, the small component can no longer be ignored.

TABLE III. Direct ionization cross sections for U^{82+}.

Subshell	Ionization potential (keV)	Cross section at twice threshold (10^{-24} cm^2)
	Hartree-Fock	
$2p$	24.81	177.2
$2s$	25.30	46.2
	Hartree-Fock relativistic	
$2p$	26.41	132.9
$2s$	29.86	26.7
	Dirac-Fock	
$2p(3/2)$	25.31	129.6
$2p(1/2)$	29.27	47.7
$2s(1/2)$	29.95	35.7

In Fig. 2 the HF, HFR, and DF total ionization cross sections for U^{82+} are presented. Only the sum of the $2p$ and $2s$ subshell direct ionization cross sections are plotted, since excitation-autoionization contributions are assumed to be small due to strong radiation field damping (i.e., $B_j^a \approx 0$ for all j).

The derivation of the product matrix elements Ω in both Eqs. (1) and (2) is based on the two-body electrostatic interaction between incident and target electrons.[14] As seen in the ionization of U^{82+}, the incident energies are becoming appreciable fractions of the rest mass energy of

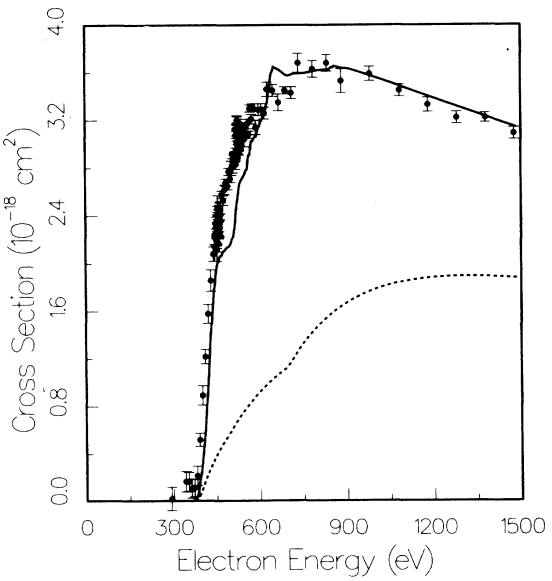

FIG. 1. Electron-impact ionization of U^{16+}. The solid curve is the total ionization cross section in the HFR approximation, the dashed curve is the direct ionization cross section only, and the experimental measurements are from Ref. 19.

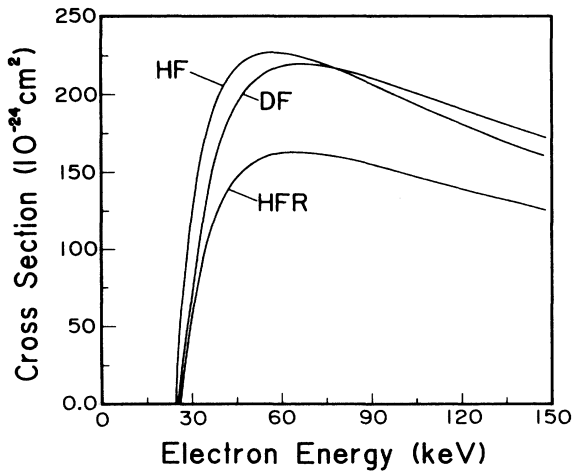

FIG. 2. Electron-impact ionization of U^{82+}. The three solid curves are different approximations for the $2s$ and $2p$ subshell direct ionization cross section: Hartree-Fock (HF), Hartree-Fock with relativistic modifications (HFR), and Dirac-Fock (DF).

the electron. At these energies the effect of the two-body magnetic interaction between incident and target electrons may have to be considered.

IV. SUMMARY

Due to the inclusive nature of the total ionization cross section for electron-ion scattering, relativistic effects are relatively small in the threshold energy region for even the heaviest atomic systems. As shown in our calculations for U^{16+}, it is much more important to keep track of indirect ionization processes, like excitation-autoionization, since they may lead to substantial enhancements of the direct ionization cross section. Exceptions to this general rule may be the K- and L-shell ionization of heavy atoms and ions. Even though our HF and DF calculations for U^{82+} are in fairly good agreement, neglected two-body magnetic interactions may change that result. Even stronger effects are predicted for K-shell ionization in the threshold energy region. In the future we hope to extend our triple partial-wave expansion calculations to include the two-body relativistic interactions needed to make more accurate predictions for K- and L-shell ionization of heavy atomic ions.

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