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Empirical model for the electron-impact K -shell-ionization cross sections

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The total cross sections of electron-impact single- K -shell ionization of 14 atomic targets ranging from H to U ($1 \leq Z \leq 92$) are calculated using a modified version of the BELI formula [Bell *et al.*, J. Phys. Chem. Ref. Data **12**, 891 (1983)] by incorporating both ionic and relativistic corrections in it. The proposed modified Bell model with a single set of parameters is found to provide an excellent description of the experimental data in the reduced energy range $1 \leq E/I_K \leq 10^6$ (E and I_K are, respectively, the incident energy and ionization potential) with a performance level at least as good as any of the existing methods and models.

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I. INTRODUCTION

The subject of K -shell ionization by electron impact (EI) has importance in fundamental studies as well in practical applications. EI K -shell-ionization cross sections are needed in a number of applied fields such as fusion plasma, radiation physics, astrophysics, etc. Reliable values of these cross sections are also required over a wide range of energies in quantitative elemental analyses using (i) electron probe microanalysis (EPMA), (ii) Auger electron spectroscopy (AES), and (iii) electron energy loss spectroscopy (EELS). For example, using EPMA, determination of thickness and elemental composition of stratified layers of thickness of the nanometer order is possible [1]. Using EPMA, AES, and EELS, the presence of pollutants, even in microscopic amounts, can be known.

A vast variety of theoretical treatments for the EI K -shell-ionization cross sections have been made. Some treatments are based upon classical mechanics and some use quantum mechanics. Each theoretical treatment has some domain validity with respect to the ranges of species and incident energies. None has been fully successful in the description of K -shell-ionization cross sections (KSIC's) over a wide range of atomic number Z and incident energies. Gryzinski's classical model [2], which has historical importance, gives fairly good agreement with a wide range of data but shows poor performance near the threshold ($U < 4$). Here U is the reduced energy defined as the ratio of incident energy E to K -shell-ionization potential I_K .

Quantum-mechanical calculations based upon the plane-wave Born approximation (PWBA) have been attempted [3–7] for the description of K -shell ionization. Luo and Joy [8] performed an extensive series of calculations of inner-shell-ionization cross sections using first-order perturbation

theory and the Hartee-Slater-Fock wave function. Scofield [9] developed an *ab initio* calculation by using the relativistic PWBA and solving the Dirac equation. Segui *et al.* [10] reported the use of the distorted-wave Born approximation (DWBA) for the K -shell-ionization cross sections with its validity in a limited energy range. Each of the theories mentioned above has a domain of limited validity and none has been found to cover wide ranges of incident energies and atomic numbers. In general, methods based upon quantum mechanics are rather difficult to implement, require a large amount of computing time, and moreover do not lead to analytical formulas for immediate use. Because of these, a large number of semiempirical and empirical models are normally used. These models have the advantage of being useful in algorithms for applications—e.g., microanalysis. These analytical models play an important role in many practical applications. Similar to quantum-mechanical calculations, each model appears to have some region of validity. For example, the semiempirical model of Green and Cosslett [11] remains valid at lower incident energies ($1 < U < 3$) but it does not produce good fits to the cross sections at higher incident energies. Quarles' model [12] extended up to five orders of U works well for some limited atoms. The empirical formula Casnati *et al.* [13] provides good description of data over the ranges $1 < U < 20$ and $6 < Z < 79$. Hombourg's model [14] gives fairly good fits to the K -shell data in the ranges $1 \leq U < 10^5$ and $6 \leq Z < 79$. Uddin *et al.* [15] proposed an improved binary-encounter dipole model with relativistic and ionic corrections (RQIBED) [16] and applied it with considerable success for the description of the K -shell-ionization of atoms in the range $6 < Z < 50$. The model with constant values of its two parameters in its structure provides an excellent description of the experimental EI cross sections even up to Sn and incident energies around 2 MeV. However, the model does not work well for ultrarelativistic incident energies beyond 10 MeV.

Bell *et al.* [17] proposed an analytical formula known in the literature as the 'Belfast ionization' (BELI) formula [18]

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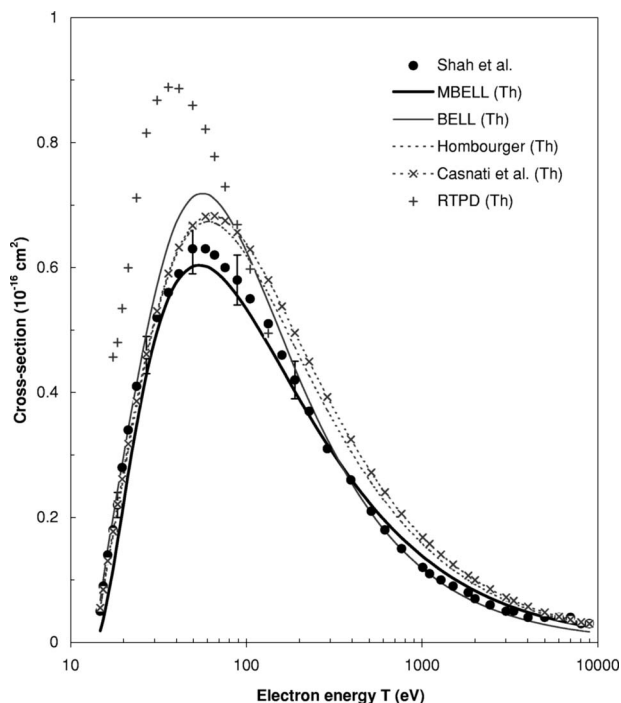


FIG. 1. Electron-impact K -shell-ionization cross sections for H. Solid circles are the experimental data from [23]. The thin solid curve, thick solid curve, dashed curve with crosses, dashed line, dashed line with open diamonds, and pluses are, respectively, the BELL [17], present MBELL calculations, results using the empirical models of Casnati *et al.* [13] and Hombourger [14], and the RTPD predictions of Kuo and Huang [55].

for the electron-impact ionization of atoms and ions and applied the formula to light atoms and ions with the parameters of the formula being species dependent. Godunov and Ivanov [18] applied the BELI formula to the EI ionization of Ne^{q+} ions. Here also no generality as to parameters of the formula was shown. In the present work, we modify the BELI formula to make it suitable for a description of the EI K -shell ionization of atoms. The BELI formula does not make any allowance for relativistic effects. We propose a modification of the BELI form to take into account relativistic and ionic effects. The model, so framed, is henceforth referred to as the modified BELI (MBELL) model, with the BELI formula herein as BELL. We apply the MBELL model to the determination of EI K -shell-ionization cross sections of H, He, C, Al, Ar, Ni, Cu, Se, Ag, Sn, Au, Pb, Bi, and U. Our predicted cross sections are compared with the available experimental results and the calculations from other models and quantum-mechanical methods.

The paper is organized as follows. The deduction of the MBELL model is outlined in Sec. II. In Sec. III, we first optimize the parameters of the MBELL model and then discuss the MBELL results for the above-mentioned 14 atoms from H to U in comparison with the available experimental and other theoretical results. Section IV is devoted to the conclusions arrived at.

II. OUTLINE OF THE MBELL MODEL

The BELL formula [17,18] for the electron-impact ionization cross sections is

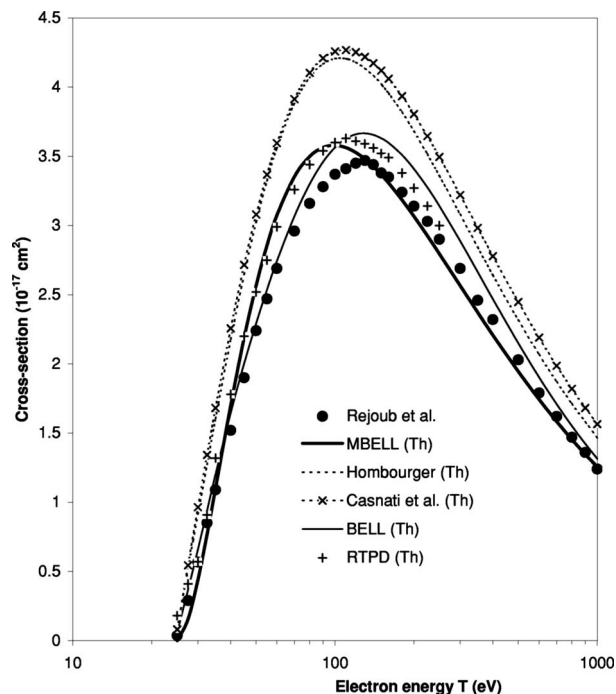


FIG. 2. Same as in Fig. 1 for He. The experimental data are solid circles from [24].

$$\sigma_{BELL}(E) = \frac{1}{I_K E} \left[A \ln \left(\frac{E}{I_K} \right) + \sum_{K=1}^5 B_K \left(1 - \frac{I_K}{E} \right)^K \right]. \quad (1)$$

As mentioned earlier, E is the energy of the incident electron, I_K is the ionization potential of the K shell, and A and B_K are the fitting coefficients. The formula is consistent with the classical scaling law [18]

$$I_K^2 \sigma(E) = \sigma_C \left(\frac{E}{I_K} \right), \quad (2)$$

assuming a Z -dependent universal function $\sigma_C(x)$ as well as the asymptotic Bethe behavior [19]

$$\sigma(E) \sim \frac{1}{I_K E} (A \ln E + B) \quad (3)$$

at asymptotic energies. The additional terms in Eq. (1) have been chosen to vanish at the threshold and to influence, through the $1/E$ term, the cross sections at intermediate energies.

In the relativistic domain of incident energies, a description of the EI ionization in the K shell of medium and heavy atoms requires relativistic treatment. We combine the relativistic factor of the Gryzinski model [2] as a multiplying factor, with the BELL form to account for the relativistic effect. The Gryzinski's relativistic factor G_R is given by

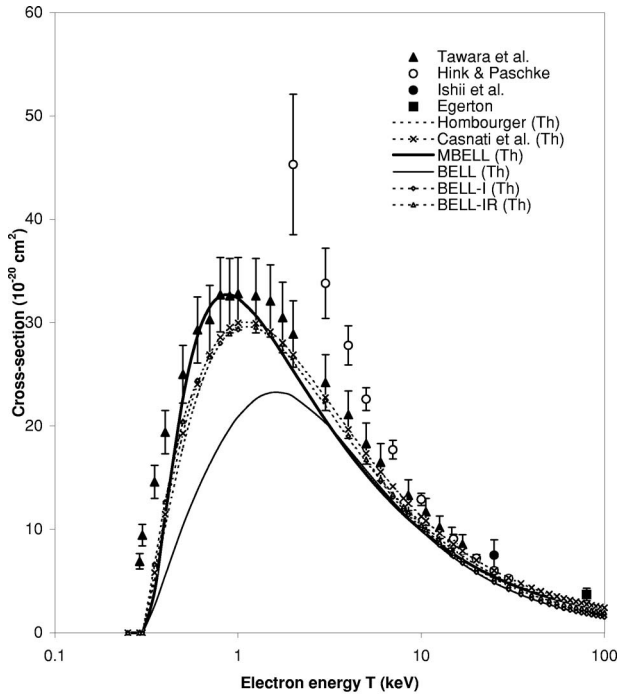


FIG. 3. Same as in Fig. 1 for C. The experimental data are from [25–28]. The dashed curve and dashed curve with crosses denote, respectively, the calculations from the empirical models of [13] and [14]. The shaded curve, dashed curve open diamonds, and dashed curve with open triangles denote, respectively, the predicted results from BELL, BELL with ionic corrections, and BELL with both the ionic and relativistic corrections.

$$G_R = \left(\frac{1+2J}{U+2J} \right) \left(\frac{U+J}{1+J} \right)^2 \times \left(\frac{(1+U)(U+2J)(1+J)^2}{J^2(1+2J)+U(U+2J)(1+J)^2} \right)^{1.5}, \quad (4)$$

with $J=mc^2/I_K$ and $U=E/I_K$. The resulting intermediate model is

$$\sigma_R(E) = G_R \sigma_{BELL}(E). \quad (5)$$

To the incident electron approaching the K -shell electron, the atom appears as an ion of charge $q=Z-N_K$ where N_K represents the number of electrons in the K shell. Thus the charge cloud of the electron is attracted towards the K shell electron, thereby leading to a greater overlap of the charge clouds of the incident and target electrons and consequent enhancement of the ionization cross section. However, the ionic effect on the cross section decreases with an increase of the incident energy as the electron spends less time in the vicinity of the field of atom. We suggest an ionic factor F_{ion} which increases in value with an increase of the charge q but decreases with the incident energy. The F_{ion} factor, in line with the form in Fontes *et al.* [20] and Uddin *et al.* [21], is taken as

$$F_{ion} = 1 + m \left(\frac{q}{ZU} \right)^n. \quad (6)$$

Here m and n are fitting parameters. The optimum values obtained for m and n , as will be discussed in Sec. III, are

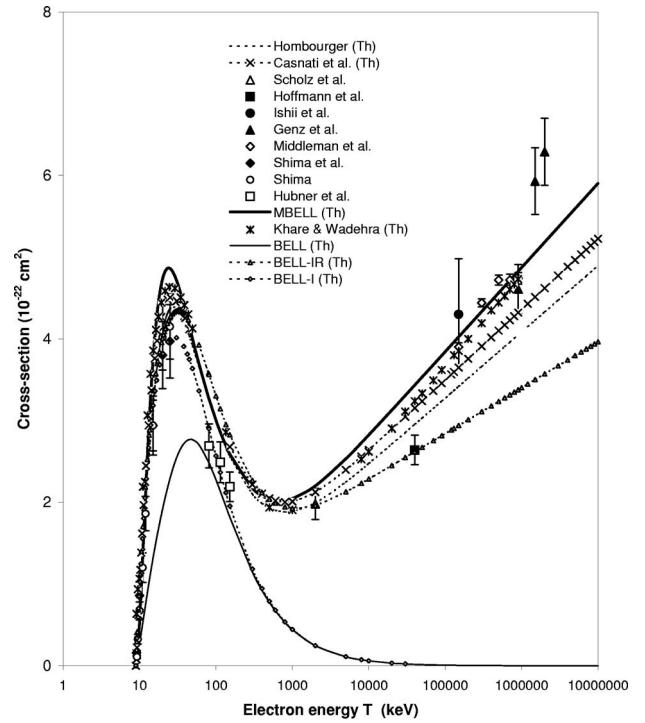


FIG. 4. Same as in Fig. 3 for Cu. The experimental data are from [27,29–37]. Asterisks are the PWBA calculations of Khare and Wadehra [7].

$m=3.00$ and $n=1.27$. We then combine the F_{ion} factor with the $\sigma_R(E)$ in Eq. (5) and add the multiplying factor N_K . The resulting cross section then becomes

$$\sigma_{MBELL}(E) = N_K F_{ion} G_R \sigma_{BELL}(E). \quad (7)$$

σ_{MBELL} now represents the EI ionization cross section for the K shell in the proposed MBELL model.

III. RESULTS AND DISCUSSIONS

The ionization potentials I_K of the K -shell electrons are taken from Desclaux [22]. The BELL formula in (1) has been found to account for well the EI cross section data of Shah *et al.* [23] for H (Fig. 1) except around the peak region and of Rejoub *et al.* [24] for He (Fig. 2). The parameter values for H, as used in Bell *et al.* [17], are $A=0.1845$, $B_1=-0.00186$, $B_2=0.1231$, $B_3=-0.1901$, $B_4=0.9527$, and $B_5=0.0$ in units of $10^{-13} \text{ eV}^2 \text{ cm}^2$ and those for He in the same units are $A=0.5720$, $B_1=-0.3440$, $B_2=-0.5230$, $B_3=3.4450$, $B_4=-6.8210$, and $B_5=5.5780$. For the K -shell EI ionization of other atoms, we use the parameters of He, as each of them involves a filled K shell as He. To examine how these parameters work for other atoms, we choose C, Cu, and Au of varying Z values as test cases. Figure 3 shows the EI ionization cross sections for C, predicted by BELL, using the above parameters. The calculated values greatly underestimate the experimental results of Tawara *et al.* [25], Hink *et al.* [26], Ishii *et al.* [27], and Egerton [28], except beyond 3 keV. The ionic factor F_{ion} in Eq. (6) with the parameter values $m=3.00$ and $n=1.27$ greatly improves the fit (curve

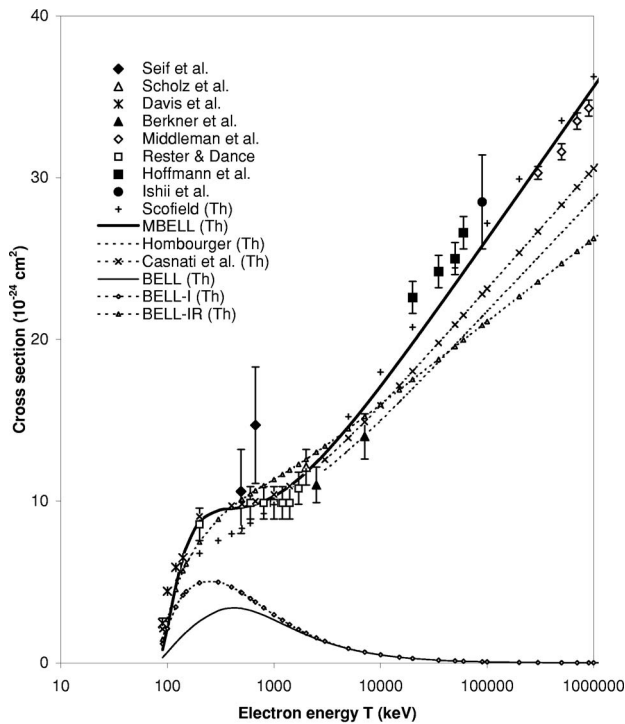


FIG. 5. Same as in Fig. 3 for Au. The experimental data are from [27,29,30,32–34,38–41]. The pluses are the relativistic PWBA predictions of [9].

labeled BELL-I in Fig. 3) to the data. The curve labeled BELL-IR denotes the BELL cross sections with both ionic and relativistic corrections. The closeness of the BELL-I and BELL-IR suggests that the relativistic factor G_R in Eq. (4)

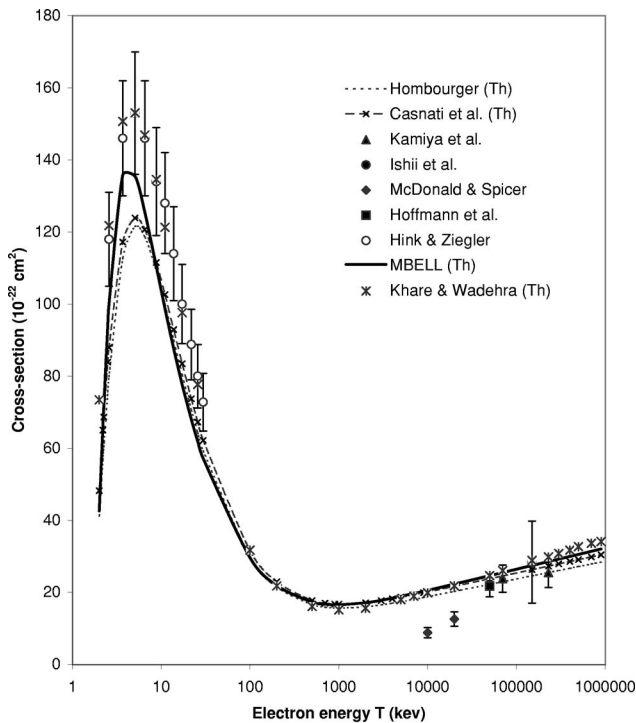


FIG. 6. Same as in Fig. 1 for Al. The experimental data are from [27,29,30,42–44]. The asterisks are the PWBA predictions of [7].

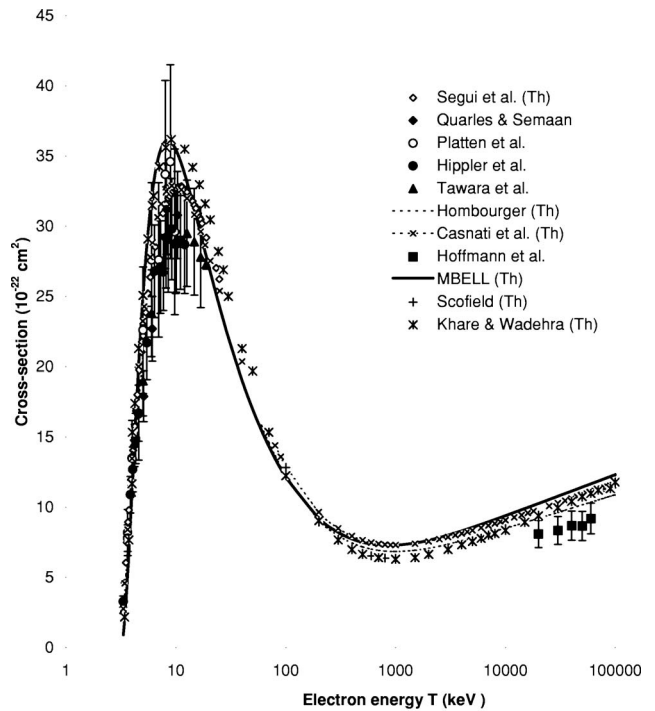


FIG. 7. Same as in Fig. 1 for Ar. The experimental data are from [25,45–47]. The asterisks, pluses, and open diamonds are, respectively, the quantal calculations of [7], [9], and [10].

seems to have a negligible effect in the energy domain considered for C. This is not unexpected as the relativistic effect is determined not only by the incident energy but also by the K -shell-ionization potential I_K . However, in the cases of Cu and Au (Figs. 4 and 5), the relativistic effects are substantial, leading to the large differences between the BELL-I and BELL-IR curves. The BELL cross sections with the parameters for He fail completely to reproduce the experimental data of Ishii *et al.* [27], Hoffmann *et al.* [29,30], Genz *et al.* [31], Scholz and co-workers [32,33], Middleman *et al.* [34], Shima *et al.* [35], Shima [36], and Hubner *et al.* [37] for Cu and of [27,29,30,32–34] as well as of Seif el Naser *et al.* [38], Davis *et al.* [39], Rester and Dance [40], and Berkner *et al.* [41] for Au. Although the BELL parameters coupled with the ionic and relativistic corrections (BELL-IR curves) greatly improve the fits in both the cases, the predicted cross sections still underestimate the data of [27,31] for Cu beyond 40 MeV and of [27,29,30,34] for Au beyond 10 MeV.

In an effort to seek an optimized set of parameter values with incorporation of ionic and relativistic corrections, we have applied the proposed MBELL model to the EI K -shell ionization of 14 targets in the range $Z=1-92$. In the MBELL model, we consider up to the fifth degree in $(1-I_K/E)$ to keep the number of parameters same as that used by Bell *et al.* [17] and Godunov and Ivanov [18]. This helps us to compare the performance of the proposed MBELL model with the parent BELL formula [17]. The sources of the experimental data, in addition to those already quoted, are Kamiya *et al.* [42], McDonald and Spicer [43], Hink and Ziegler [44], Quarles and Semaan [45], Platten *et al.* [46], Hippler *et al.* [47], Jessenberger and Hink [48], Pockman *et al.* [49], Smick and Kirkpatrick [50], Berenyi *et al.* [51], Kiss *et al.*

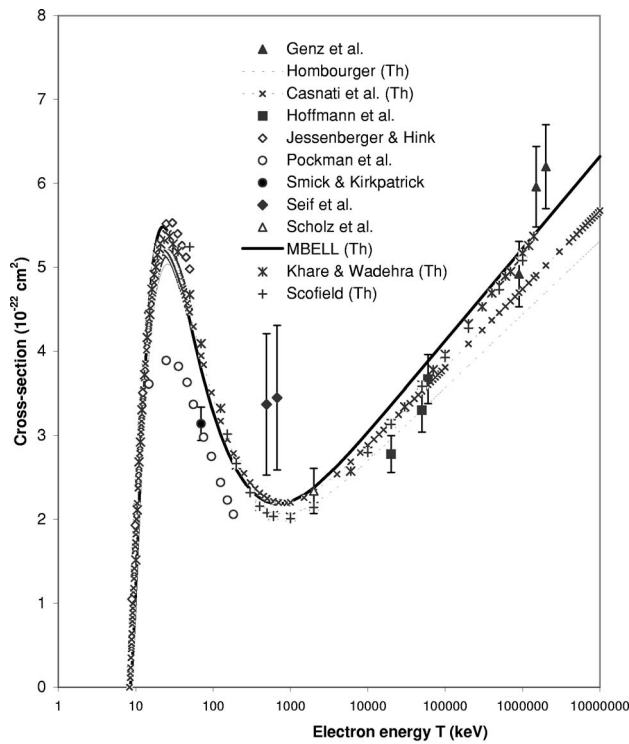


FIG. 8. Same as in Fig. 7 for Ni. The experimental data are from [29–33,38,48–50].

[52], Schlenk *et al.* [53], and Ricz *et al.* [54]. The parameters A , B_1 , B_2 , B_3 , B_4 , and B_5 are optimized to obtain best fits to the data of all the atoms. The optimum values of these parameters are found to be $A=0.525$, $B_1=-0.510$, $B_2=0.200$, $B_3=0.050$, $B_4=-0.025$, and $B_5=-0.100$ in units of $10^{-13} \text{ eV}^2 \text{ cm}^2$. This single set of values of the parameters is applied to the MBELL model for calculating the EI K -shell-ionization cross section of all atoms, including H, He, and C, considered herein.

In Figs. 1–14, the predictions of the present MBELL model, shown as thick solid curves, are compared with the available experimental data as well as the empirical calculations of Casnati *et al.* [13] and Hombourger [14], PWBA calculations of Khare and Wadehra [7], perturbation calculations (with exchange effects) of Luo and Joy [8], and relativistic PWBA calculations of Scofield [9], relativistic DWBA calculations of Segui *et al.* [10], and relativistic two-potential (RTPD) DWBA calculations of Kuo and Huang [55].

It is clearly evident from the figures that the MBELL model describes all the experimental data either excellently or satisfactorily (within 10%–15%) except the data of [31] beyond 1500 MeV for Cu (Fig. 4) and the data of [34] beyond 300 MeV for Bi (Fig. 13). The MBELL predictions compare closely with the results of the quantum-mechanical calculations of Kuo and Huang [55] for He (Fig. 2); Khare and Wadehra [7] for Cu (Fig. 4), Al, Ar, Ni, Se (Figs. 6–9), and Sn (Fig. 11); Scofield [9] for Au (Fig. 5), Ar, Ni (Figs. 7 and 8), Ag (Fig. 10), and Bi except around the 500-keV energy region (Fig. 13); and Segui *et al.* [10] for Ar (Fig. 7) and Ag except around 100 keV region (Fig. 10). The MBELL model describes the experimental data of H much

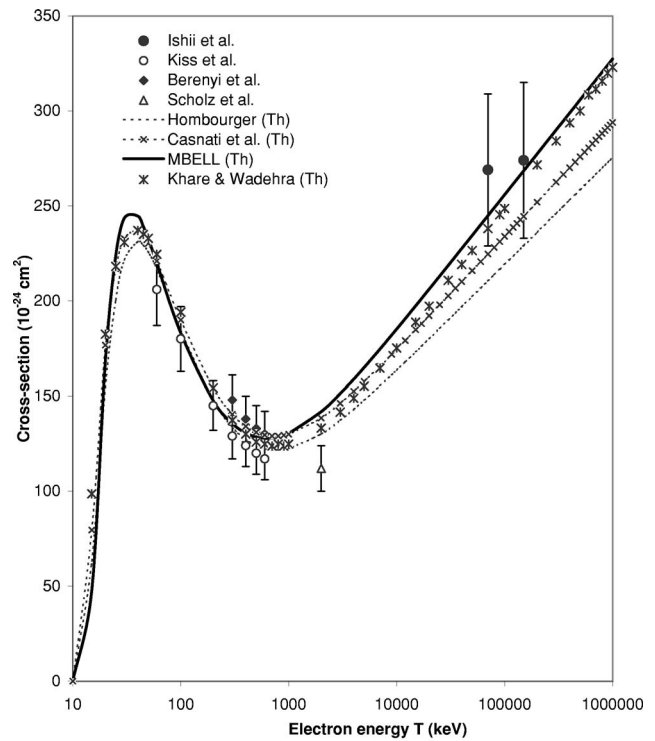


FIG. 9. Same as in Fig. 7 for Se. The experimental data are from [27,32,33,51,52].

better than even the quantal RTPD method [55]. The overall performance of the MBELL model is better than that of the empirical models of Casnati *et al.* [13] and Hombourger [14].

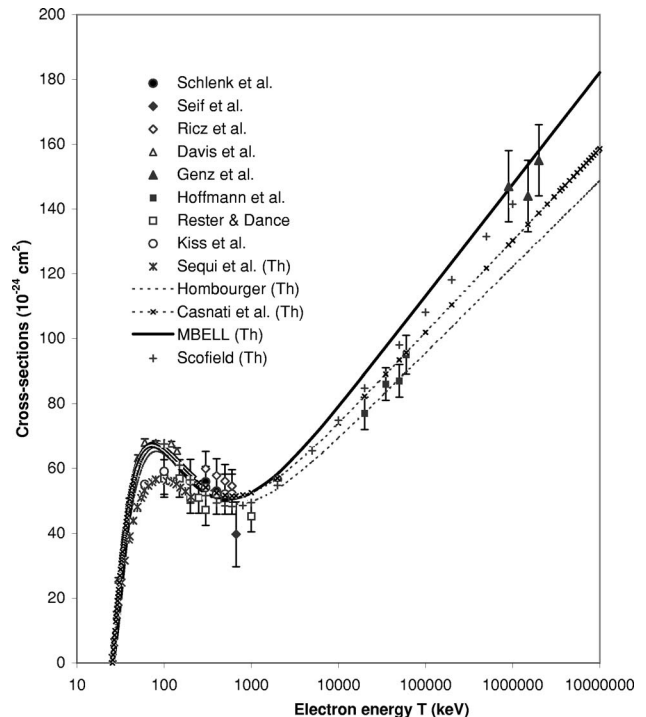


FIG. 10. Same as in Fig. 7 for Ag. The experimental data are from [29–31,38–40,52–54].

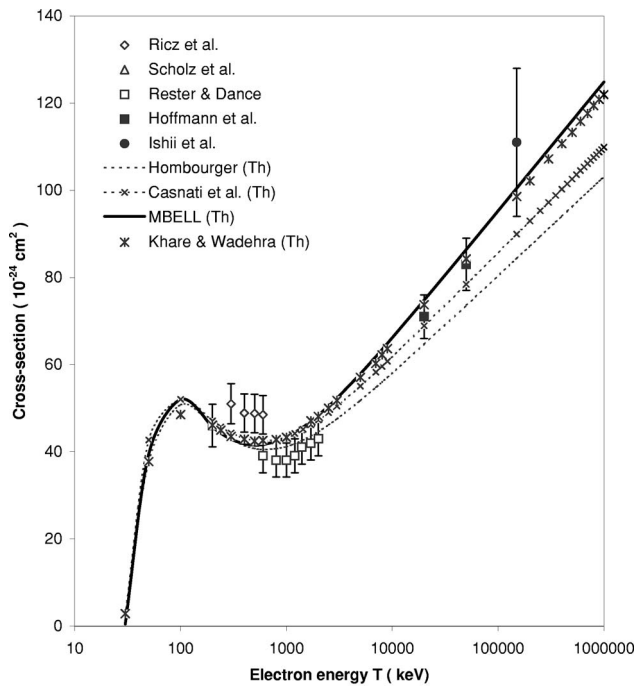


FIG. 11. Same as in Fig. 7 for Sn. The experimental data are from [27,29,30,32,33,40,54].

IV. CONCLUSIONS

The present MBELL model, with a single set of values for the parameters over the targets considered in the range $1 \leq Z \leq 92$ and the reduced incident energy range $1 \leq U \leq 10^6$, seems to be the best overall performer, with respect to the experimental cross sections, among the theoretical methods

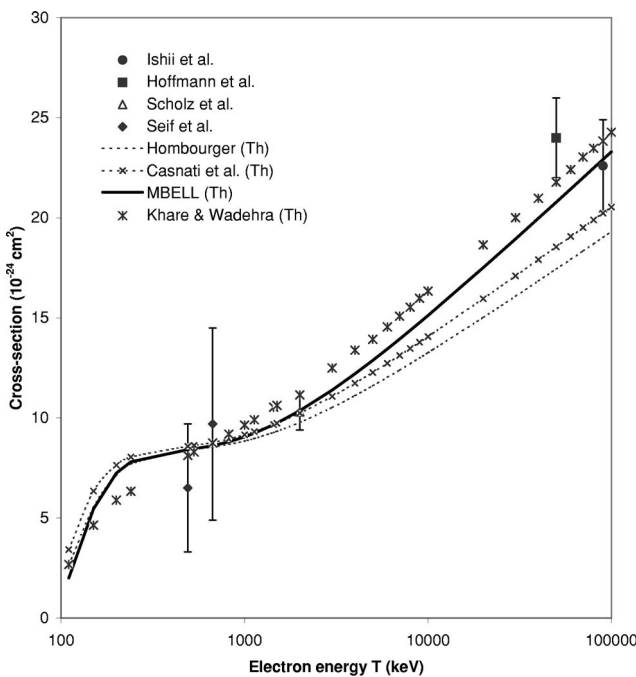


FIG. 12. Same as in Fig. 7 for Pb. The experimental data are from [27,29,30,32,33,38].

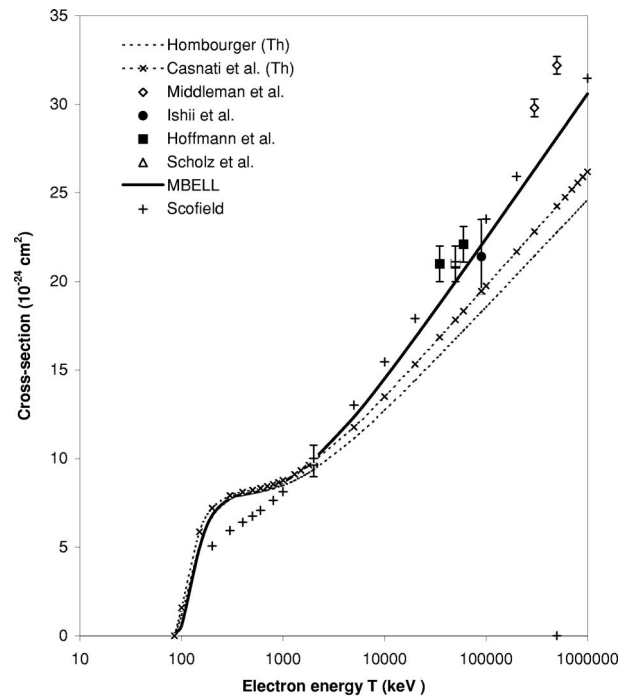


FIG. 13. Same as in Fig. 7 for Bi. The experimental data are from [27,29,30,32–34].

and models considered. As far as we know, the ranges of both Z and U in the present study are extended beyond the available empirical and quantal calculations, except for those due to Khare and Wadehra [7]. However, for light targets the threshold energy is very low and hence in this region the

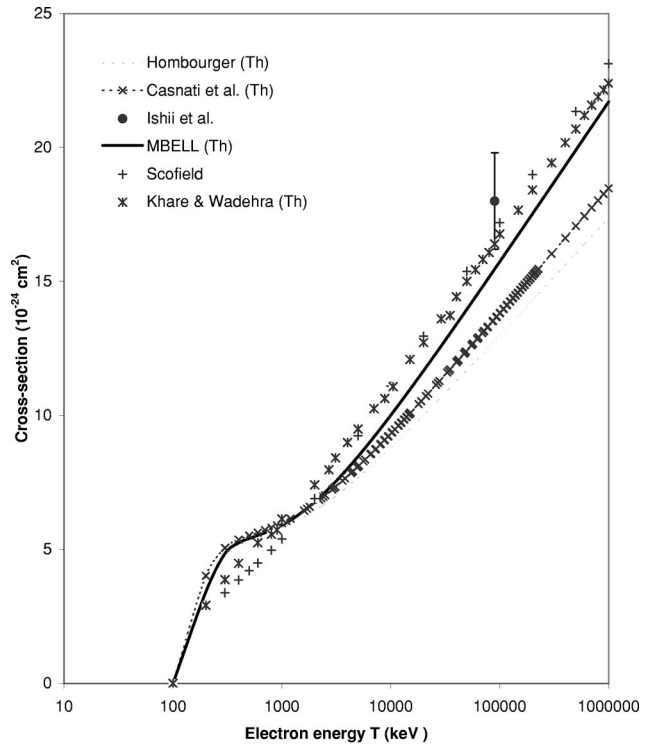


FIG. 14. Same as in Fig. 7 for U. The experimental data are from [27].

PWBA results may not be reliable since the Born approximation remains valid at higher energies. The PWBA method entails a prior knowledge of the generalized oscillator strength, whose accurate determination is essential for obtaining the correct result and involves numerical integration including the oscillator strength. Our proposed MBELL model is very simple to implement and sufficiently accurate, as demonstrated in this study, over not only wide ranges of incident energies but also various atomic species. Consider-

ing the overall performance of the present MBELL model, we anticipate that it may become a very useful model for future applications.

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