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The effect of the relativistic Hartree–Fock–Roothaan momentum distribution in the binary encounter approximation for inner shell ionization

V KUMAR, S N CHATTERJEE and B N ROY
Department of Physics, Bihar University, Muzaffarpur 842001, India

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Abstract. K- and L-shell ionization cross sections of gold due to impact of proton and alpha particles have been calculated in the binary encounter approximation incorporating the effects of Coulomb deflection of the projectile, of increase in binding of the target electron, and of polarization and relativity Relativistic Hartree–Fock–Roothaan (RHFR) momentum distributions for the target electron along with an approximate relativistic correction to the collision dynamics have been used in the present calculations A comparison with the corresponding calculations using non-relativistic Hartree–Fock–Roothaan wave functions, experimental results and other available calculations is presented.

Keywords. Alpha particle, proton; K-shell ionization; L-shell ionization; relativistic

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1. Introduction

Ionization from inner shells of atoms, particularly K and L shells due to impact of charged particles has been a subject of intensive study because of its wide applications (Hardt and Watson 1976; Starter *et al* 1981). A systematic study of experimental and theoretical cross sections for light ion impact K-shell ionization of different atoms are available in literature (e.g. Paul and Obermann 1983; Paul 1984, 1987, 1989). In this context the two review articles, Paul (1984) and Paul and Muhr (1986), are important. The experimental cross sections available in literature have been systematically studied for proton impact L-shell ionization of atoms, in the recent past by Braziewicz and Braziewicz (1988). These studies indicate that the ion impact inner shell ionization process is best described in the intermediate energy region by ECPSSR method which is a modified version of plane wave born approximation (PWBA) including corrections for the energy loss (E), Coulomb deflection (C), perturbed stationary state effect (PSS) and the relativistic effect (R). However, a direct calculation by this method is involved and requires much computation. On the other hand the binary encounter approximation (BEA) has been successfully used for a theoretical investigation of these processes with comparatively much small computer time (for example Chatterjee *et al* 1983; Srivastava *et al* 1984; Shrivastava and Roy 1986). In fact the BEA, apart from its simplicity, is essentially linked with the plane wave Born approximation (PWBA) and gives results which are in agreement with PWBA (Langenberg 1978). However, in most of the previous BEA calculations on inner-shell ionization cross sections by charged particle impact of atoms the relativistic nature of the target electron has been ignored. But this must be taken into account especially for heavy targets. For heavy targets the relativistic effects have been found to increase the cross sections significantly, particularly at low impact energy (Avaldi *et al* 1982). Avaldi *et al* (1982) have reported their BEA calculations on proton impact K-shell ionization cross section for Au. They have incorporated the effect of relativity using relativistic collision dynamics and a relativistic hydrogenic momentum distribution for the bound electron. Sheth (1984) has reported BEA calculations on proton impact K-shell ionization of Ti and Cu incorporating relativistic effects for the bound electron following a different approach. He has used the relativistic version of the Rutherford cross section up to the first order Born approximation, incorporating the correction due to relativity and spin along with the Dirac wave function. Although, the use of hydrogenic functions is approximately correct in K-shell electrons, it is not at all physically justified for other shells. We have used RHFR wave functions for both K and L shell electrons in the present calculations.

In the present work, Vrien's (1967) expressions for the ionization cross section of an atom due to impact of heavy charged particles have been used. The effects of Coulomb repulsion of the projectile due to the target nucleus and of the change in binding of the target electron due to presence of the projectile have been incorporated following the method of Thomas and Garcia (1969) and Brandt and Lapicki (1979) respectively. In order to account for relativistic effects of the target electron, approximate relativistic collision dynamics introduced by Avaldi *et al* (1982) has been used along with the RHFR momentum distribution function for the bound electron which have been developed as suggested by Mukoyama and Kagawa (1983).

2. Theoretical consideration

Using atomic units, with energy expressed in rydbergs, Vriens's expression (1967) for ionization cross section due to impact of a heavy charged particle, incorporating the modification for Coulomb repulsion of the projectile and change in the binding energy of the target electron may be written as

$$Q=\begin{cases}\dfrac{(s+s')^2 Z_1^2}{s^2 s'^2 U_c^2\, m}\left\{1+\dfrac{2}{3}mt^2-\dfrac{1}{4m(s'^2-t^2)}\right\}(\pi a_0^2); & 1\leqslant 4ms'(s'-t)\\[2ex] \dfrac{(s+s')^2}{2s^2 s'^2 U_c^2 tm}Z_1^2\left[\dfrac{1}{4m(s'+t)}+t+\dfrac{2}{3}m\left\{2s'^3+t^3-\left(\dfrac{1}{m}+t^2\right)^{3/2}\right\}\right](\pi a_0^2); & 4ms'(s'-t)\leqslant 1\leqslant 4ms'(s'+t)\\[2ex] 0; & 1\geqslant ms'(s'+t)\end{cases} \quad (1)$$

where

$$t^2=v_{2s}^2/U_c,\quad s^2=v_1^2/U_c$$

$$s'^2=s^2-(1{\cdot}058Z_1Z_{2s})/(1836Ma_{2s}U_c),$$

with v_1 and v_{2s} (subscript s stand for K, L_1, L_2, L_3) the velocity of the projectile and bound electron, respectively, Z_1 and M, the charge and mass (in units of proton mass) of the projectile, a_{2s} the radius of the shell considered, $Z_{2s}=Z_2-S_{2s}$ with Z_2 the nuclear charge of the target and S_{2s} the screening constant of the shell, and m the ratio of relativistic to non-relativistic mass of the electron. In (1) $U_c = \varepsilon U_{2s}$ is the binding energy of the target electron corrected for the presence of the projectile whereas U_{2s} is the unperturbed binding energy of the target electron. The correction factor ε (Brandt and Lapicki 1979), incorporating the effects of increase in binding energy of the target electron and the polarization of the orbital, is given by

$$\varepsilon = 1 + (2Z_1/Z_{2s}\theta_{2s}) \quad (g - h),$$

with $\theta_{2s} = (U_{2s}/Z_{2s}^2)$ (13·6), and, for K shell:

$$g = (1 + 5x + 7{\cdot}14x^2 + 4{\cdot}27x^3 + 0{\cdot}947x^4)/(1 + x)^5,$$

for L_1 shell:

$$g = (1 + 9x + 31x^2 + 49x^3 + 162x^4 + 63x^5 + 18x^6 + 1{\cdot}97x^7)/(1 + x)^9,$$

and for L_2 and L_3 sub shells:

$$g = (1 + 10x + 45x^2 + 102x^3 + 331x^4 + 6{\cdot}7x^5 + 58x^6 + 7{\cdot}8x^7 + 0{\cdot}888x^8)/(1 + x)^{10}$$

where $x = v_1/(\frac{1}{2}\theta_{2s}v_{2s})$,

$$h = (2n_2/\theta_{2s}x^3)I(C_s n_2/x),$$

$$I(\kappa) = 2\exp(-2\kappa)/\kappa^{1{\cdot}6},$$

$$C_K = C_{L_1} = 3/2 \text{ and } C_{L_2} = C_{L_3} = 5/4.$$

Cross sections have been obtained by integrating the above expressions over the Hartree-Fock-Roothaan (HFR) and RHFR momentum distribution for the target electron. In principle, for relativistic considerations, the upper limit of the velocity of the target electron should be c, the speed of light in vacuum. However, the contributions from large values of t (and hence v_{2s} the velocity of the target electron) are negligibly small. Hence during the course of evaluting the integrals over the momentum distribution we have used a cut off for t determined by a fixed accuracy limit of the cross sections, in order to obtain convergence of the integral. The HFR momentum distribution for the target electron has been constructed using the double zeta function taken from the table of Mclean and Mclean (1981).

Here we would like to present a brief discussion on the method of constructing RHFR momentum distribution function for the target electron. In the configuration space the RHFR wave function is given as (Kagawa 1975)

$$\psi_{s\kappa u}(\gamma) = \frac{1}{\gamma}\begin{pmatrix} P_{s\kappa}(\gamma) & \chi_\kappa^u(\gamma) \\ iQ_{s\kappa}(\gamma) & \chi_{-\kappa}^u(\gamma) \end{pmatrix} \qquad (2)$$

where $P_{s\kappa}(\gamma)$ and $Q_{s\kappa}(\gamma)$ are the large and small components of the radial wave function, $\chi_\kappa^u(\hat{\gamma})$ is the spin angular wave function and S is the principal quantum number.

The relativistic quantum number κ is expressed as $\kappa = \pm(j + \frac{1}{2})$ for $j = l \pm \frac{1}{2}$, where j is the total angular momentum and l is the orbital angular momentum. The radial wave functions are given in terms of the STO with non-integral principal quantum number

$$P_{s\kappa}(\gamma)=\sum_q \xi_{sq}^{(l)} f_{\kappa q}(\gamma) \tag{3}$$

and

$$Q_{s\kappa}(\gamma)=\sum_q \xi_{sq}^{(s)} f_{\kappa q}(\gamma). \tag{4}$$

Here $\xi_{sq}^{(l)}$ and $\xi_{sq}^{(s)}$ are the expansion coefficients.

The STO $f_{\kappa q}(\gamma)$ is given by

$$f_{\kappa q}(\gamma)=(2\zeta_{\kappa q})^{n'_{\kappa q}+\frac{1}{2}}[\Gamma(2n'_{\kappa q}+1)]^{-1/2}\gamma^{n'_{\kappa q}}\exp(-\zeta_{\kappa q}\gamma) \tag{5}$$

where

$$n'_{\kappa q}=n_{\kappa q}+(\kappa^2-z^2\alpha^2) \quad n_{\kappa q}=0,1,\dots. \tag{6}$$

and $\zeta_{\kappa q}, \Gamma(x), z$ and α are orbital exponent, gamma function, atomic number and fine structure constant, respectively.

The momentum representation of the wave function of (2) is given by (Rubinowicz 1948)

$$\psi_{s\kappa u}(p)=\begin{pmatrix} N_{s\kappa}(p) & \chi_\kappa^u(p) \\ iM_{s\kappa}(p) & \chi_{\bar{\kappa}}^u(p)\end{pmatrix}. \tag{7}$$

The radial wave functions are obtained from those in the configuration space through the Fourier Bessel transformation

$$N_{s\kappa}(p)=i^{-l}\left(\frac{2}{\lambda}\right)^{1/2}\int_0^\infty j_l(p\gamma)P_{s\kappa}(\gamma)\,\mathrm{d}\gamma \tag{8a}$$

$$M_{s\kappa}(p)=i^{-\bar{l}}\left(\frac{2}{\lambda}\right)^{1/2}\int_0^\infty j_{\bar{l}}(p\gamma)Q_{s\kappa}(\gamma)\,\mathrm{d}\gamma \tag{8b}$$

where $j_l(x)$ is the spherical Bessel function of the first kind and $\bar{l}$ denotes the orbital. angular momentum corresponds to $-\kappa$.

Using (3) and (4), eqs. (8a) and (8b) reduce to

$$N_{s\kappa}(p)=i^{-l}\sum_q \xi_{sq}^{(l)} C_{\kappa q} I(n'_{\kappa q},\zeta_{\kappa q},l) \tag{9a}$$

$$M_{s\kappa}(p)=i^{-\bar{l}}\sum_q \xi_{sq}^{(s)} C_{\kappa q} I(n'_{\kappa q},\zeta_{\kappa q},\bar{l}) \tag{9b}$$

where

$$C_{\kappa q}=(2\zeta_{\kappa q})^{n'_{\kappa q}+(1/2)}/[\Gamma(2n^1_{\kappa q}+1)]^{1/2} \tag{10}$$

and

$$I(n,\zeta,l)=\frac{\Gamma(m)p^l}{2^{l+2}\zeta^m\Gamma(l+\frac{3}{2})}F\left(\frac{m}{2},\frac{m+1}{2};l+\tfrac{3}{2};\frac{-p^2}{\zeta^2}\right)$$

with $m=n+l+2$.

Finally the RHFR momentum distribution for an orbital is given by $P(p)=|N_{s\kappa}(p)|^2+|M_{s\kappa}(p)|^2$ (see Mukoyama and Kagawa 1983). For these calculations, parameters made available by Mukoyama (private communication) have been used. These parameters were made available by Mukoyama only for gold and hence it was not possible to extend the calculations for other systems in the present work. The binding energies of the target electron and the quantum mechanical values of the screening constant have been taken from Lotz (1968) and Fischer (1972) respectively. For the shell radii of atoms the quantum mechanical values of points of maximum radial probability density reported by Desclaux (1973) have been used.

3. Results and discussion

We have calculated proton and alpha particle impact K-shell and L-shell ionization cross sections of gold following the method outlined in § 2. Two sets of calculations have been carried out for each projectile. The results have been presented in the figures. In the first set of calculations the effects of the Coulomb repulsion of the projectile, the increase in the binding energy of the target electron and polarization effects have been incorporated along with a non-relativistic HFR wave functions for the bound electron. In the second set of calculations the effects of relativity have also been incorporated. The calculated results have been compared with available experimental observations and other theoretical calculations, particularly the ECPSSR originally formulated by Brandt and Lapicki (1979).

The present results for proton and alpha particle impact K-shell ionization cross sections have been shown in figures 1 and 2 respectively. The BEA calculations for proton impact by Avaldi *et al* (1982) overestimate the cross sections. This is as expected because they have used a relativistic hydrogenic wave function for the bound electron. In the present work the target electron has been described by a RHFR wave function.

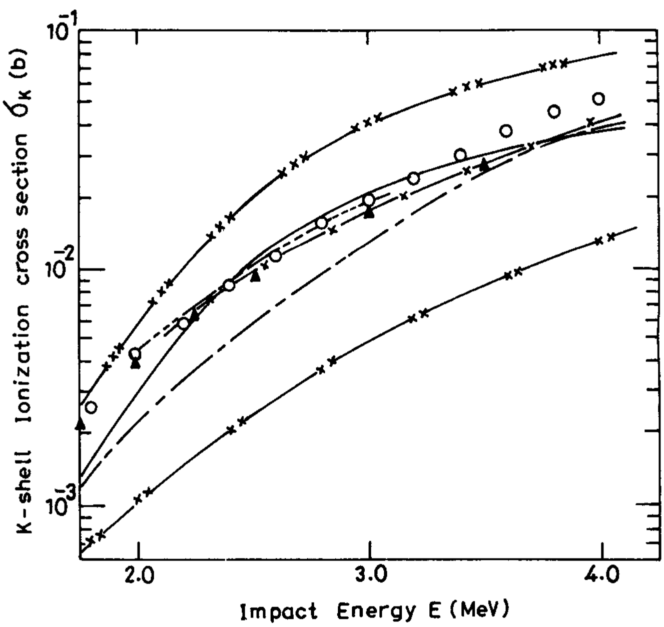

Figure 1. —K-shell ionization cross sections for H^+ impact. Experimental data: O, Kamiya *et al* (1977) Δ, Castro Faria *et al* (1984). Theoretical curves: —— × × × ———, BEA with relativistic hydrogenic wave functions (Avaldi *et al* (1982) —— × × ——, Present non-relativistic results; ——— ——— ——, PWBA with electronic relativistic wave functions (Jamnik and Zupanic 1957), ——————, Present relativistic results, —— × ——, ECPSSR Theory (Cohen and Harrigan 1985) ———— – – – ——— RPWBA-BC (Chen and Crasemann 1985).

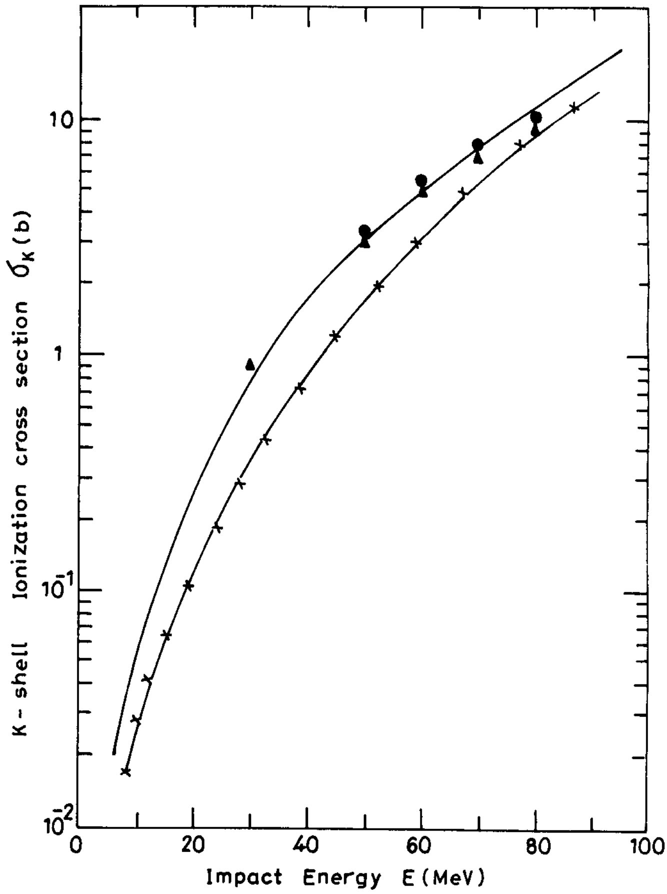

Figure 2. K-shell ionization cross section for He^{2+} impact. Experimental data ● and Δ, Hardt and Watson (1973) – × – × –, present results incorporating effects of relativistic collision dynamics only (see text); ———, present relativistic results.

It has been found that the use of a Hartree-Fock wave function lowers the cross sections, especially at low impact energies (Kumar and Roy 1981). Further, the present results are closer to experiments as compared to relativistic PWBA calculations of Jamnik and Zupancic (1957). It is also seen from figure 1 that relativistic treatment of the collision and use of a relativistic momentum function for the bound electron increases the cross sections, leading to a closer agreement with experiment. To examine the effects of relativistic treatment of collision dynamics on cross sections, we have carried out calculations for alpha particle impact (figure 2), using relativistic wave functions only. As expected, relativistic consideration of collision gives rise to a significant increase in cross section. The present results for alpha particle impact including all corrections are in close agreement with other experimental and theoretical results. Figures 3 and 4 show the results for L_1 sub-shell. The present calculations for proton impact (incorporating all physical effects) overestimate most of the experimental cross sections throughout the energy range investigated. However, different sets of experimental results show significant differences among themselves and hence more experimental and theoretical investigations are needed to understand the reason

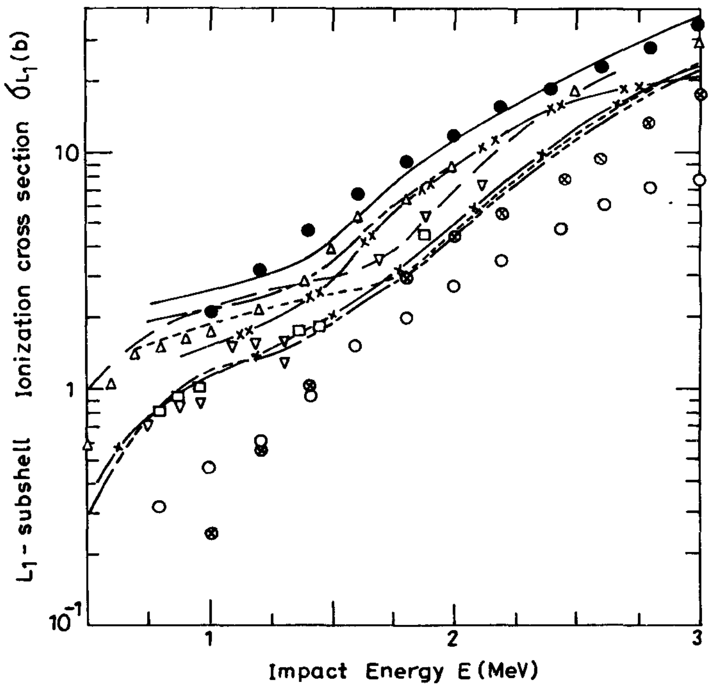

Figure 3. L_1 subshell ionization cross sections for H^+ impact. Experimental data: ○ and ⊗, Cohen (1980); □, Chen (1977); ∇, Datz *et al* (1974); ● and Δ, Cohen and Pinho respectively (reported by Sokhi and Crumpton (1984)). Theoretical curves: – – –, ———, calculations in PWBA, and PWBAR respectively (reported by Cohen 1980); – – – – – –, PWBA-DHS (Chen 1984).—— × × ——, present non-relativistic results; ————, Present relativistic results, – – – – – – – –, RPWBA-BC (Chen and Crasemann 1985)— × —ECPSSR theory (Cohen and Harrigan 1985).

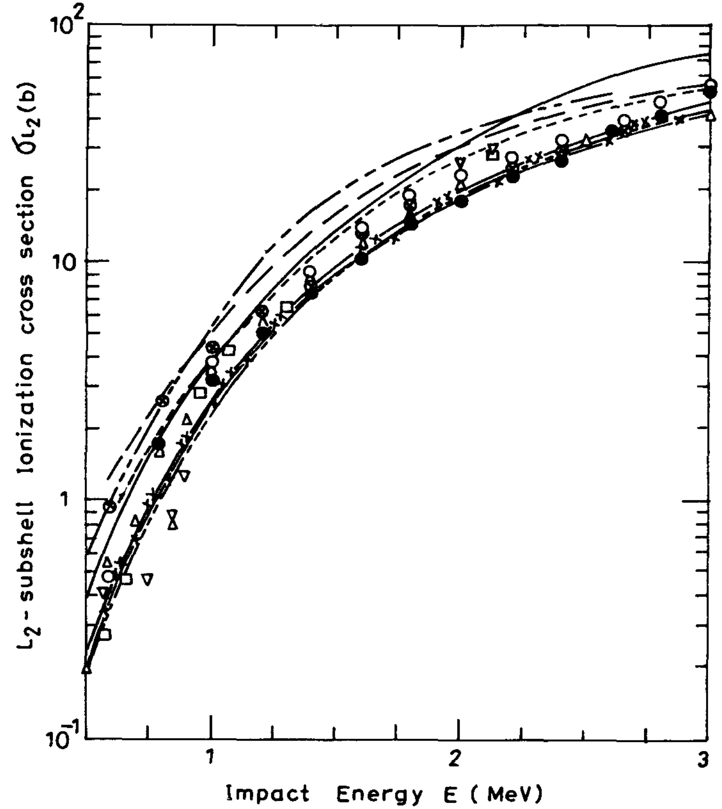

Figure 4. L_1 Subshell ionization cross sections for He^{2+} impact – – – × – – –, present non-relativistic result, ———— present relativistic results; ——— × ———, ECPSSR theory (Cohen and Harrigan 1985) Experimental ●, Chang *et al* (1975) reported by Hardt and Watson (1976).

behind this discrepancy. In fact all the available theoretical calculations (shown in figure 3) overestimate the experimental cross sections considerably. As observed in experiments, an inflexion has been obtained in the present cross section curve which has been attributed to the presence of an extra node in the L_1 sub-shell wave function. The position of the inflexion in our work is close to the experimental counterpart. As is seen from the figure, results of theoretical calculations using different approximations differ significantly. The present results are closer to the recent calculations of Chen (1984) in the relativistic plane wave Born approximation (RPWBA) using relativistic Hartree-Slater (RHS) wave function for the target electron. For alpha particle impact (Figure 4) also our calculations show good agreement with other results. From the figure it is clear that, as expected, incorporation of relativity in the calculation enhances the cross sections considerably.

Cross sections for L_2 sub-shell have been presented in figures 5 and 6. The present results for proton impact are in reasonable agreement with the experimental cross sections (figure 5). Our results also show agreement with other theoretical calculations. Recent relativistic calculations in RPWBA along with RHS wave function for the bound electron reported by Chen (1984) are larger than experimental as well as theoretical cross sections at low impact energies; however, with increase in impact energy RPWBA results improve. Our results for alpha particle impact (figure 6) have been compared with the theoretical results in ECPSSR reported by Cohen and Harrigan (1985) and experimental data of Chang *et al* (1975). Present calculations are larger than experimental as well as other theoretical cross sections.

The present results for proton impact L_3 sub-shell ionization cross sections (figure 7) are within a factor of 2 as compared to the experimental cross sections throughout

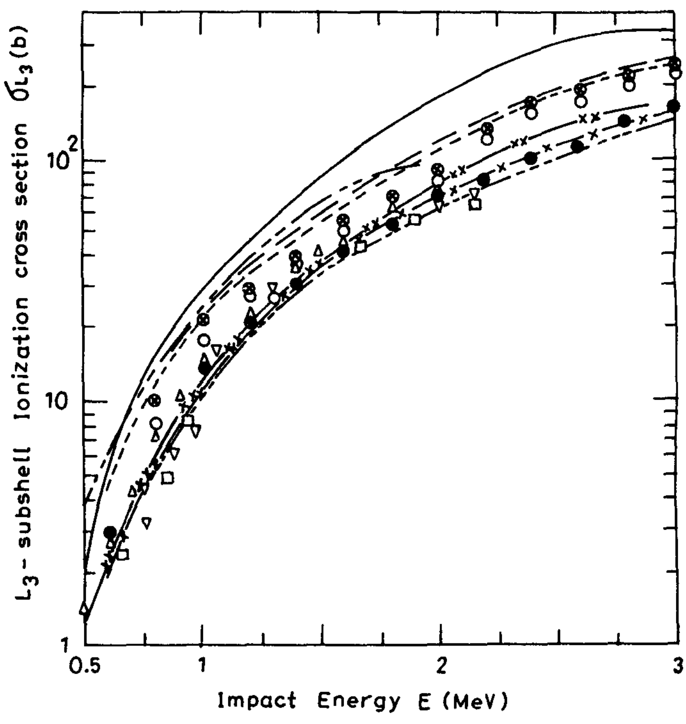

Figure 5. L_2 subshell ionization cross sections for H^+ impact. Same as in fig 3.

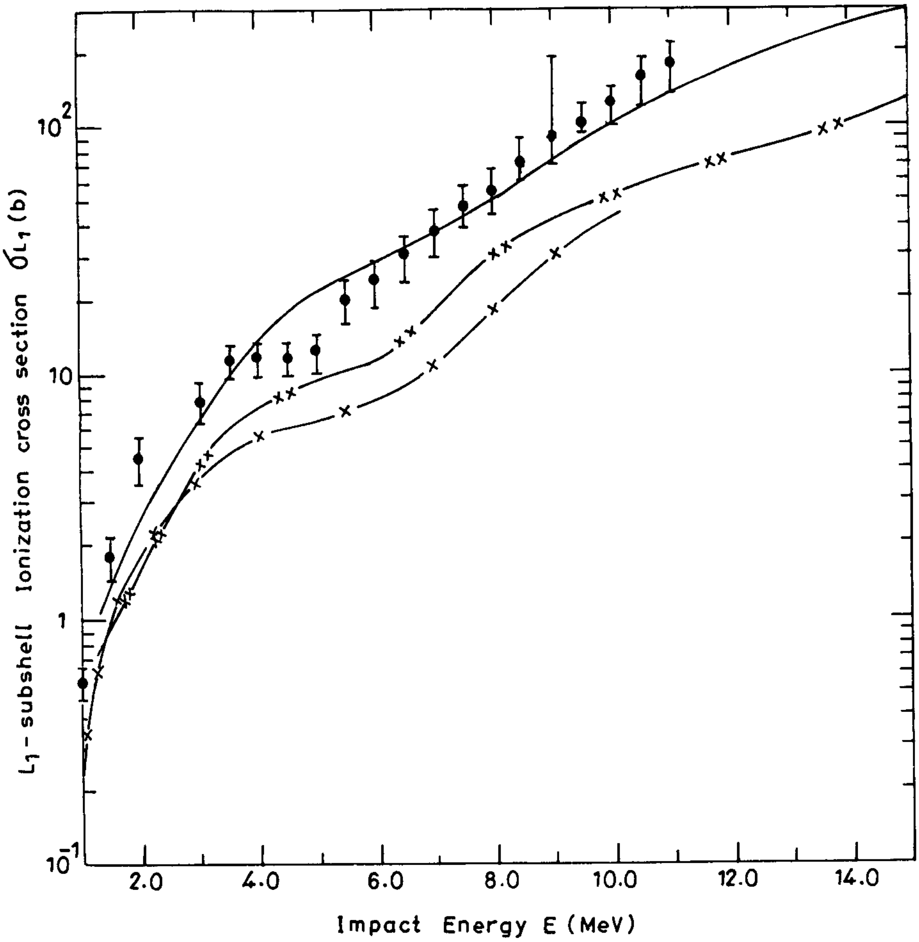

Figure 6. L_2 subshell ionization cross sections for He^{2+} impact Same as in figure 4

the energy range investigated. Except the recent RPWBA calculation of Chen (1984) using RHS wave function for the target electron, all theoretical calculations show almost similar type of variation of cross sections with energy, in agreement with experiments.

From our calculated cross sections we have obtained the values $\sigma_{L_1}/\sigma_{L_2}$ and $\sigma_{L_1}/\sigma_{L_3}$ for proton and alpha particle impact and have compared them with the available experimental observations and other theoretical calculations (Figures 9–12). The proton impact experimental values of these ratios, obtained by different workers, show marked differences among themselves. The present calculations of $\sigma_{L_1}/\sigma_{L_2}$ are slightly higher than the results of ECPSSR reported by Cohen and Harrigan (1985) but in $\sigma_{L_1}/\sigma_{L_3}$ the present values are in close agreement with other results. Our results for $\sigma_{L_1}/\sigma_{L_2}$ and $\sigma_{L_1}/\sigma_{L_3}$ in He^{2+} impact show good agreement with the experimental observations and other theoretical calculations. In fact the present results are always within a factor of 2 as compared to experimental values.

From a close inspection of the figures it is concluded that the present method, in spite of being simple, gives satisfactory description of the relativistic effects for heavy particle impact inner shell ionization of atoms. Our results, in most cases, are higher

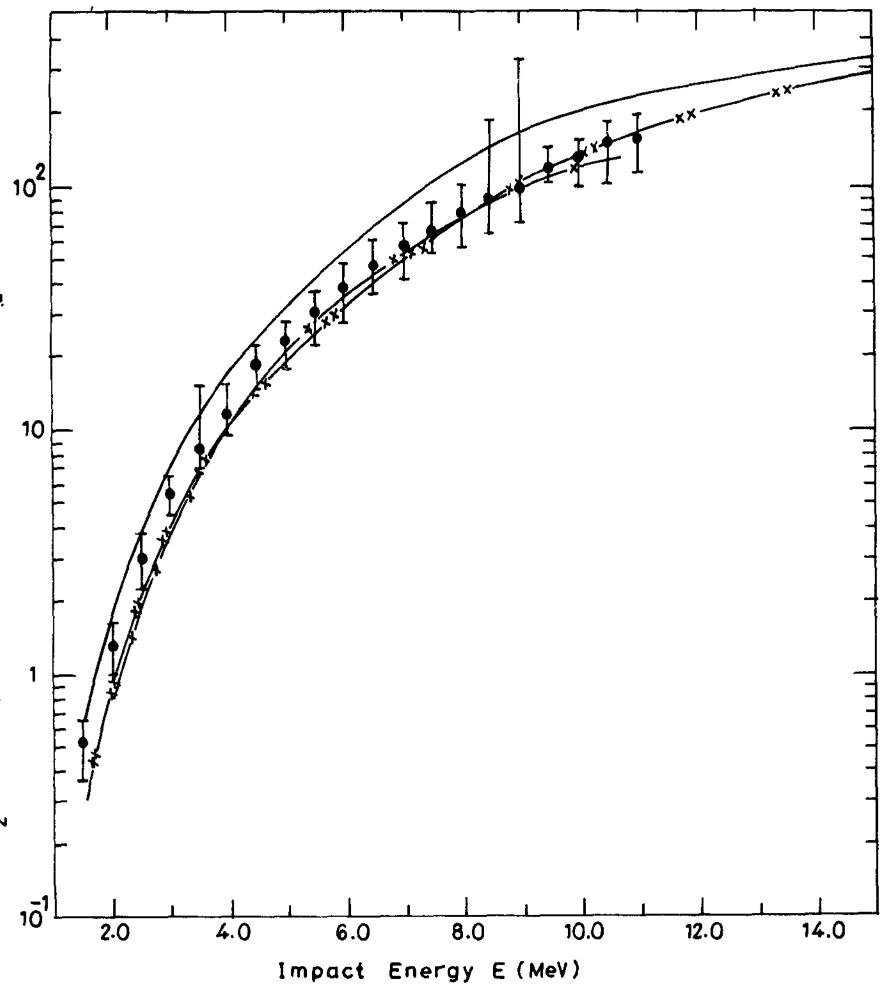

Figure 7. L_3 subshell ionization cross sections for H^+ impact. Same as in figure 3.

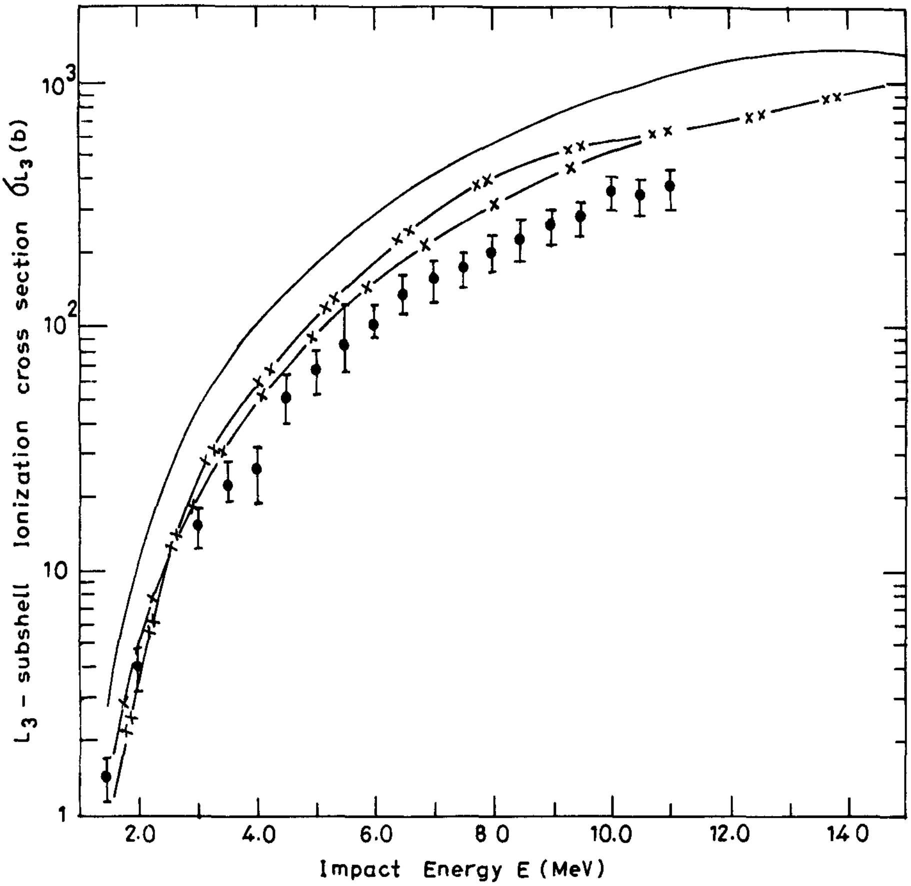

Figure 8. L_3 subshell ionization cross sections for He^{2+} impact. Same as in figure 4.

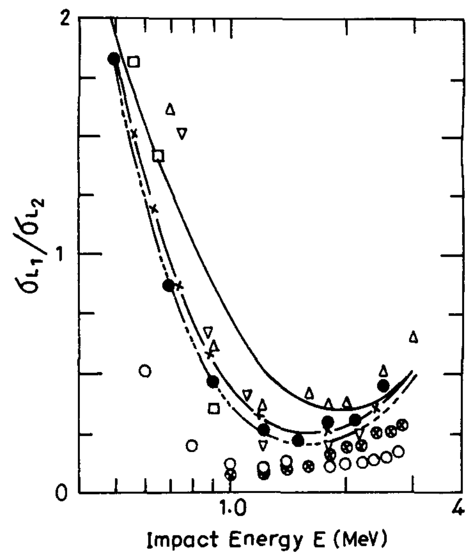

Figure 9. Ionization cross section ratio $\sigma_{L_1}/\sigma_{L_2}$ for H^+ impact. Experimental data: ●, Jitchin *et al* (1982); □, Chen (1977); ∇, Datz *et al* (1974); ○ and × Thin and Thick target measurements (Cohen 1980). Δ, Sokhi and Crumpton (1984). Theoretical curves: ————, Present relativistic results; —— × ——, ECPSSR Theory (Cohen and Harrigan 1985), ——— – – ———, RPWBA-BC (Chen and Crasemann 1985).

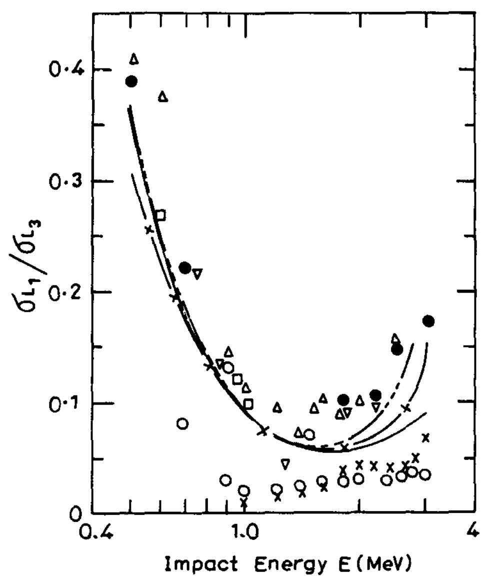

Figure 10. Ionization cross section ratio $\sigma_{L_1}/\sigma_{L_3}$ for H^+ impact Same as in figure 9

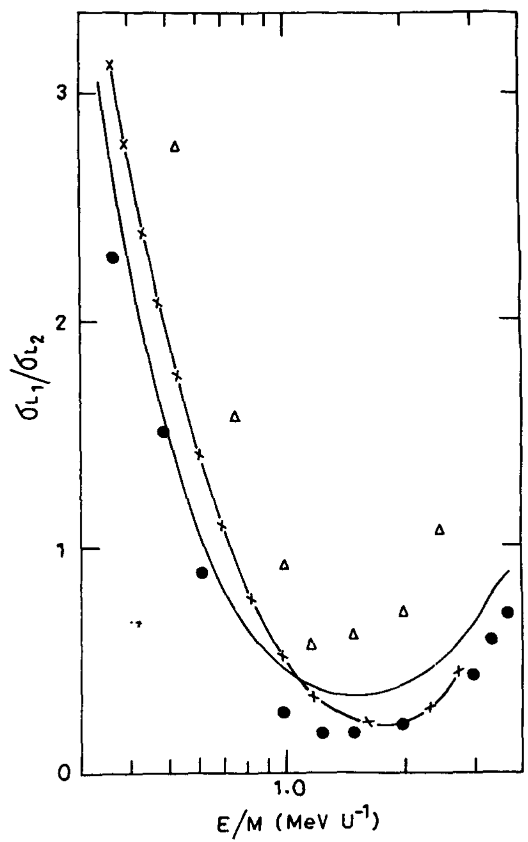

Figure 11. Ionization cross section ratio $\sigma_{L_1}/\sigma_{L_2}$ for He^{2+} impact. ●, Experimental data (Jitchin *et al* 1983); Δ, Hardt and Watson (1976). Theoretical curves ———, present relativistic results. ——×——, ECPSSR Theory (Cohen and Harrigan 1985).

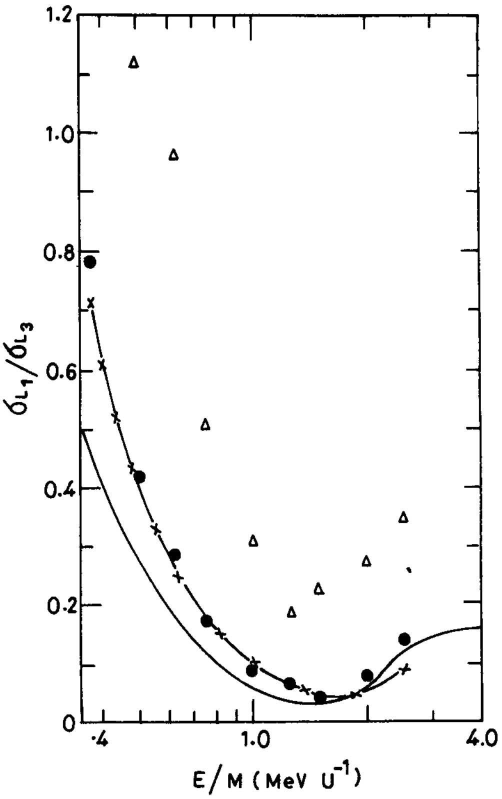

Figure 12. Ionization cross section ratio $\sigma_{L_1}/\sigma_{L_3}$ for He^{2+} impact. Same as in figure 11

than the cross sections obtained experimentally or calculated using different quantal approximations, particularly at low impact energies. This might be due to the use of the binary encounter approximation in the present work.

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