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He$^+$ Impact Double Ionization of Noble Gases

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ABSTRACT Theoretical calculations of He$^+$ impact double ionization of Ne, Ar, Kr and Xe have been performed in the Modified Binary Encounter Approximation. Direct double ionization cross sections have been calculated in the modified binary encounter model. Accurate expression of $\sigma_{\Delta E}$ (cross section for energy transfer ΔE) and Hatree-Fock velocity distributions for the target electrons have been used throughout the calculations. The present results of double ionization cross sections are in excellent agreement with the experimental observations in the case of Ne, Ar and Kr throughout the energy range. The calculated cross sections differs from the experimental results in the low energy regions in case of Xe because the present approximation not exhibits better result in the low energy regions, while the over-estimations of experimental results in the high energy regions shows that more theoretical calculations is required to understand the dynamics of the system.

KEYWORDS Hartree-Fock, Double Ionization, Cross-sections, Vriens
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INTRODUCTION

The understanding of interaction of heavy particles with atoms and ions are atomic processes of fundamental nature. H$^+$ impact ionization plays a significant role not only in different fields of Physics, but also in other branches of science. Single ionization is usually the most important among various ionization processes, but multiple ionization (especially double ionization) is strongest in various environments with abundance of energetic electrons. When compared with different multiple ionization processes, double ionization (DI) has the largest impact on ionization state distribution. The theoretical and experimental studies of double ionization have been widely accepted [1-4]. Direct and Indirect processes are responsible for the formation of the charge state of the resulting ion with two removed electrons. Indirect process is determined by ionization and subsequent auto-ionization.

On the other hand, direct process occurs due to simultaneous ejection of two electrons from the target to continuum.

Direct double ionization is attracting the researchers for considerable interest due to complex nature of four-body Coulomb potential where correlation effects are of the largest importance. Many theoretical methods [5-6] show a good agreement with experimental measurements for the two electron system. It should be mentioned that only direct processes take place in such systems. Furthermore, time-dependent close coupling approach has been successfully used to analyze systems with more than two electrons [7-8]. However, these studies include the systems with one or two electrons outside the core and are totally ineffective for more complex systems due to very rigorous calculations. As per our knowledge, there are no approaches which would provide a good agreement with experimental data for such atoms.

Two step model works very well for indirect process when initial ionization leads to additional removal of electron due to auto-ionization. It is gradually accepted that direct double ionization can't be described by sequential ionization because of strong correlated motion of electrons. Exception is the shake-off process which takes place after initial ionization at high electron energies. Hence sophisticated calculations of the total double ionization cross sections of many electron atoms by ion impact are not available in the literature. However, in the past, the Binary Encounter Approximation (BEA) has been used successfully in calculations of charged particle impact single and double ionization cross sections for several atoms and ions.

Gryzinski [1] reasonably considered two processes in a double Binary Encounter Model to describe double ionization. In the first process, the two electrons may be ejected from the system by two successive interactions of the incident particle with the target electrons. Next, the incident particle may knock out only one target electron and the second electron is removed by the first ejected electron. The corresponding cross sections are denoted by Q_{sc}^{ii} (scattered part) and Q_{ej}^{ii} (ejected part) respectively. The idea of two-step interaction has been supported by a number of workers [9-10]. Later on, the results of double ionization cross sections based on the modified model including contributions of indirect physical processes were found to be in close agreement with the experimental data [11-12]. In these calculations Hatree-Fock (HF) and hydrogenic velocity distributions were used while considering ejection of the first and the second target electron respectively. Later on, Jha and Roy [13-14] used HF velocity distribution while considering the ejection of both electrons of the target in calculations of direct double ionization cross sections.

In case of heavy charged particle the Binary Encounter calculations of double ionization cross sections of atoms are scarce in literature. Kumar and Roy [15] pointed out errors and obscurities in Gryzinski's theory for calculations of the above mentioned processes and modified the mathematical framework suitably incorporating the necessary corrections of using the accurate expression of $\sigma_{\Delta E}$ (cross section for energy transfer ΔE) as given by Vriens [16]. They calculated proton impact double ionization cross sections of noble gases [15-17] which were found to be in satisfactory agreement with the experimental observations. In these calculations they have used HF and hydrogenic velocity distributions for considering the ejection of the first and the second electron respectively. Later on, Singh et al [18] have used HF velocity distribution function for considering the ejection of both the electrons respectively in case of H^+ and He^+ impact double ionization of Mg and found satisfactory agreement with the experiment.

Keeping the above mentioned facts in view, we have considered it worthwhile to carry out calculation of He^+ impact double ionization of cross sections of Ne, Ar, Kr and Xe using HF velocity distribution for all the ejected electrons. This work will enable us to analyze single and direct double ionization cross sections and to examine the contributions to double ionization from indirect physical processes.

THEORETICAL DETAILS

In accordance with the prediction of the first Born Approximation, the single ionization cross-section depends on charge Z of the incoming particle and its velocity V as $Z^2 V^{-2} \ln V$, if the velocity is much larger than that corresponding to the binding energy of the atomic electron [19]. Here we like to mention that we have assumed Z^2 dependence also in calculations of direct double ionization cross section in the present double Binary Encounter Model justification of which will be given after the presentation of the mathematical expressions. In the present work, we have used the accurate expressions of cross-sections as given by Vriens [16] for heavy charged particles incident on atoms. Following the theory of Catlow and McDowel [20] we have defined two dimensionless variables s and t defined by

$$s^2 = \frac{v_1^2}{v_0^2} \text{ and } t^2 = \frac{v_2^2}{v_0^2}$$

where v_1 and v_2 are the velocities in the atomic units of the incident particle and the target electron respectively and $u = v_0^2$ is the ionization potential of the target in Rydberg. Entire energies involved have also been expressed in Rydbergs. In terms of these dimensionless variables, the expressions of ionization cross-sections due to projectile of unit charge for particular incident energy and a particular velocity of a bound electron were given by (see Kumar and Roy [15]).

$$Q_i(s, t) = \frac{4}{s^2u^2}\left[1 + \frac{2t^2}{3} - \frac{1}{4(s^2-t^2)}\right], \quad 1 \le 4s(s-t)$$

$$= \frac{2}{s^2u^2t}\left[\frac{1}{4(s+t)} + t + \frac{2}{3}\left\{2s^3 + t^3 - (1+t^2)^{3/2}\right\}\right], \quad 4s(s-t) \le 1 \le 4s(s+t) \qquad (1)$$

$$= 0, \quad 1 > 4s(s+t).$$

Numerical integration of the expression for $Q_i(s,t)$ has been carried out over Hartree-Fock velocity distribution of the bound electron to obtain the ionization cross-section. Thus, the expression of heavy charged particle impact single ionization cross-section for a particular shell of the target is given by

$$Q_i(s) = n_e Z^2 \int_0^\infty Q_i(s,t) f(t)\, u^{1/2}\, dt\, (\pi a_0^2) \qquad (2)$$

where n_e is the number of electrons in shell, Z is the charge on the projectile and $f(t)$ is the momentum distribution function of the target electron.

Heavy charged particle impact double ionization cross section including the contribution from Auger emission can be written as

$$Q^{ii}(T) = Q_D^{ii} + Q_A^{ii} \qquad (3)$$

Although the contribution of Auger emission is not taken in the present calculation, where Q_D^{ii} denotes the contribution from direct ejection of the two electrons and Q_A^{ii} from the Auger emission. Here Q_D^{ii} is given by

$$Q_D^{ii} = Q_{sc}^{ii} + Q_{ej}^{ii}\,. \qquad (4)$$

In accordance with the idea given by Gryzinski [1] in the double Binary Encounter Model, these cross sections involving integrals over energy transfer is given by

$$Q_{sc}^{ii} = \frac{n_e(n_e-1)}{4\Pi\bar{r}^2} \int_{u_i}^{\Delta E_{max}} \sigma_{\Delta E}(E_q) \left(\int_{u_{ii}}^{\Delta E_{max}} \sigma_{\Delta E'}(E_q - \Delta E)\, d(\Delta E') \right) d(\Delta E) \qquad (5)$$

and

$$Q_{ej}^{ii} = \frac{n_e^{'}(n_e-1)}{4\Pi\bar{r}^2} \int_{u_i+u_{ii}}^{\Delta E_{max}} \sigma_{\Delta E}(E_q)\left(\int_{u_{ii}}^{\Delta E-u_i} \sigma_{\Delta E^{'}}(\Delta E)d(\Delta E^{'})\right)d(\Delta E) \qquad (6)$$

The various symbols in the above expressions have been defined by Gryzinski [1]. Here ΔE and $\Delta E^{'}$ stands for energy transfers during the first and the second collisions respectively and $\bar{r}$ denotes the mean distance between the electrons in the shell which is given by $\bar{r} = \frac{R}{n_e^{1/3}}$ (R being the radius of the shell of the target atom). u_i and u_{ii} are the ionization potentials corresponding to ejection of the electrons of the target. The symbol E_q represents the energy of the projectile.

In terms of dimensionless variables s and t discussed earlier, the expression for $\sigma_{\Delta E}$ in case of a projectile of unit charge is given by (see Kumar and Roy [15])

$$\sigma_{\Delta E}d(\Delta E) = \left\{\begin{matrix} Ad(\Delta E); & \Delta E \le 4su(s-t) \\ Bd(\Delta E); & 4su(s-t) \le \Delta E \le 4su(s+t) \\ 0\ ; & \Delta E > 4su(s+t) \end{matrix}\right\} \qquad (7)$$

where

$$A = \frac{4}{s^2u}\left(\frac{1}{(\Delta E)^2} + \frac{4t^2u}{3(\Delta E)^3}\right)$$

and

$$B = \frac{2}{3t(\Delta E)^3}\left(8s - \frac{\left[(\Delta E+t^2u)^{1/2} - tu^{1/2}\right]^3}{s^2u^{3/2}}\right).$$

The expressions of the scattered part of the direct double ionization cross sections showing the relevant integrals involving energy transfer and Hartree-Fock velocity distributions for the ejection of the two target electrons are given below

$$Q_{sc}^{ii} = \frac{n_e(n_e-1)Z^2}{4\Pi\bar{r}^2}\left(\begin{matrix} \int_{t=0}^{s-\frac{1}{4s}}\left\{\int_{u_i}^{4su_i(s-t)} A\alpha d(\Delta E) + \int_{4su_i(s-t)}^{4su_i(s+t)} B\alpha d(\Delta E)\right\} f(t)u_i^{\frac{1}{2}}dt \\ + \int_{t=s-\frac{1}{4s}}^{\infty}\int_{u_i}^{4su_i(s+t)} B\alpha f(t)u_i^{\frac{1}{2}}d(\Delta E)dt \end{matrix}\right)(\Pi a_0^2) \qquad (8)$$

when (s-1/4s) is positive
and

$$Q_{sc}^{ii} = \frac{n_e(n_e-1)Z^2}{4\Pi\bar{r}^2}\left(\int_{t=\frac{1}{4s}-s}^{\infty}\int_{u_i}^{4su_i(s+t)} B\alpha f(t)u_i^{\frac{1}{2}}d(\Delta E)dt\right)(\Pi a_0^2) \qquad (9)$$

when (s-1/4s) is negative.

In the above expressions

$$\alpha = \int_0^{\infty} Q_i(s^{'},t)f^{'}(t)u_{ii}^{\frac{1}{2}}dt(\Pi a_0^2) \qquad (10)$$

here s' is given by

$$s'^2 = \left\{\begin{matrix} \frac{E_q-\Delta E}{1836u_{ii}} & for\ H^+impact \\ \frac{E_q-\Delta E}{7344u_{ii}} & for\ He^{2+}impact \end{matrix}\right\} \qquad (11)$$

similarly the expressions for ejected part are

$$Q_{ej}^{ii} = \frac{n_e(n_e-1)Z^2}{4\Pi\bar{r}^2}\left(\begin{array}{c}\int_{t=0}^{s-(1+\frac{u_{ii}}{u_i})/4s}\left\{\int_{u_i+u_{ii}}^{4su_i(s-t)} A\alpha' d(\Delta E) + \int_{4su_i(s-t)}^{4su_i(s+t)} B\alpha' d(\Delta E)\right\} f(t)u_i^{\frac{1}{2}}dt \\ + \int_{t=s-\frac{(1+\frac{u_{ii}}{u_i})}{4s}}^{\infty} \int_{u_i+u_{ii}}^{4su_i(s+t)} B\alpha' f(t)u_i^{\frac{1}{2}}d(\Delta E)dt\end{array}\right)(\Pi a_0^2) \qquad (12)$$

when $s-(1+\frac{u_{ii}}{u_i})/4s$ is positive
and

$$Q_{ej}^{ii} = \frac{n_e(n_e-1)Z^2}{4\Pi\bar{r}^2}\left(\int_{t=\frac{\left(1+\frac{u_{ii}}{u_i}\right)}{4s}-s}^{\infty} \int_{u_i+u_{ii}}^{4su_i(s+t)} B\alpha' f(t)u_i^{\frac{1}{2}}d(\Delta E)dt\right)(\Pi a_0^2) \qquad (13)$$

when $s-(1+\frac{u_{ii}}{u_i})/4s$ is negative
with

$$\alpha' = \int_0^{\infty} q_i(s',t)f'(t)u_{ii}^{\frac{1}{2}}dt(\Pi a_0^2) \qquad (14)$$

Here $q_i(s', t)$ is the expression for electron impact ionization cross section of atoms (see Jha and Roy [13-14]) and s' is given by $s'^2 = \frac{\Delta E - u_i}{u_{ii}}$ for both H^+ and He^+ impact.

Now we discuss the Z^2 dependence of the expression of Q_{sc}^{ii} which denotes a process in which the projectile knocks out two electrons successively. In a quantum mechanical approach this corresponds to a second order process for which cross section scales as Z^4. In this connection it is pertinent to point out the observations made by Vriens [16] that the two double Binary Encounter Processes are linked with the quantum mechanical first and second order approximations. If one uses correlated many electron wave functions, direct double ionization cross section will be finite even in the first born approximation. This has been assumed to correspond to Q_{ej}^{ii} of the process of direct double ionization. There is also a contribution to direct double ionization from the second born approximation, which includes double processes like those represented by Q_{sc}^{ii}. In the present method the contributions of Q_{ej}^{ii} are found to be much smaller than those of Q_{sc}^{ii} (see also Kumar and Roy [15]. In case of proton impact Z=1 and therefore, Z^4 scaling for Q_{sc}^{ii} becomes essentially the same as Z^2 scaling and good agreement of calculated results with the experiment is achieved. However, in case of alpha particle impact, calculation involves Z=2 and a Z^4 scaling of Q_{sc}^{ii} leads to much dominant contribution of this process adversely affecting the results. Hence the correspondence of the processes represented by Q_{ej}^{ii} and Q_{sc}^{ii} to the first and the second born approximations does not appear to be suitable. In this context the experimental results of H^+ and He^+ impact pure double ionization cross sections are noteworthy. It is seen that the pure double ionization cros sections are about an order smaller than the corresponding single ionization cross sections which indicates usual trend of direct double ionization. Keeping these observations in view, we have assumed Z^2 dependence of direct double ionization cross sections in the present calculations as no established dependence of direct double ionization cross sections on Z is available for this purpose.

The integrals appearing in Q_{sc}^{ii} and Q_{ej}^{ii} have been evaluated numerically. The functions *f(t)* and *f ′(t)* appearing in the above equations are momentum distribution functions corresponding to first and the second ejected electro respectively. These have been constructed from Hartree-Fock radial wave functions (see Catlow and McDowell [20], Jha and Roy [13-14]). In order to obtain Q_A^{ii} (contribution to double ionization from Auger emission), the single ionization cross section should be multiplied by Auger yield of the shell under consideration. The factor $\frac{n_e(n_e-1)}{4\Pi\bar{r}^2}$ has been suitably modified for considering the mode of ionization in which the electrons are ejected from different shells. In this case $n_e\ (n_e\ -1)$ has been replaced by $n_{e1}\ x\ n_{e2}$, where these two stands for number of electrons in the shells under consideration.

RESULTS AND DISCUSSION

The cross sections for double ionization of noble gases by He^+ impact have been calculated in the energy range 1.0 MeV to 3.5 MeV. The present theoretical results are compared with the measured data of Santos et al [21] in the above mentioned energy range and it has been shown in the Table 1-4 and Figure 1-4 respectively.

The calculated double ionization cross sections of Ne by He^+ impact in the energy range 1.0 MeV -3.5 MeV have been shown in the Table 1 and Figure 1.

Table 1: Double ionization of Neon atom by He^+ impact in units of 10^{-18} cm^2

Energy (MeV)	2p,2p	2p,2s	total	Expt. [21]
1.00	26.37	3.17	29.54	15
1.25	20.61	2.31	22.92	
1.50	16.25	1.74	17.99	8.3
2.00	11.32	1.14	12.46	7.5
2.50	7.62	0.75	8.37	5.3
3.00	5.62	0.55	6.17	4.9
3.50	4.30	0.43	4.73	4.4

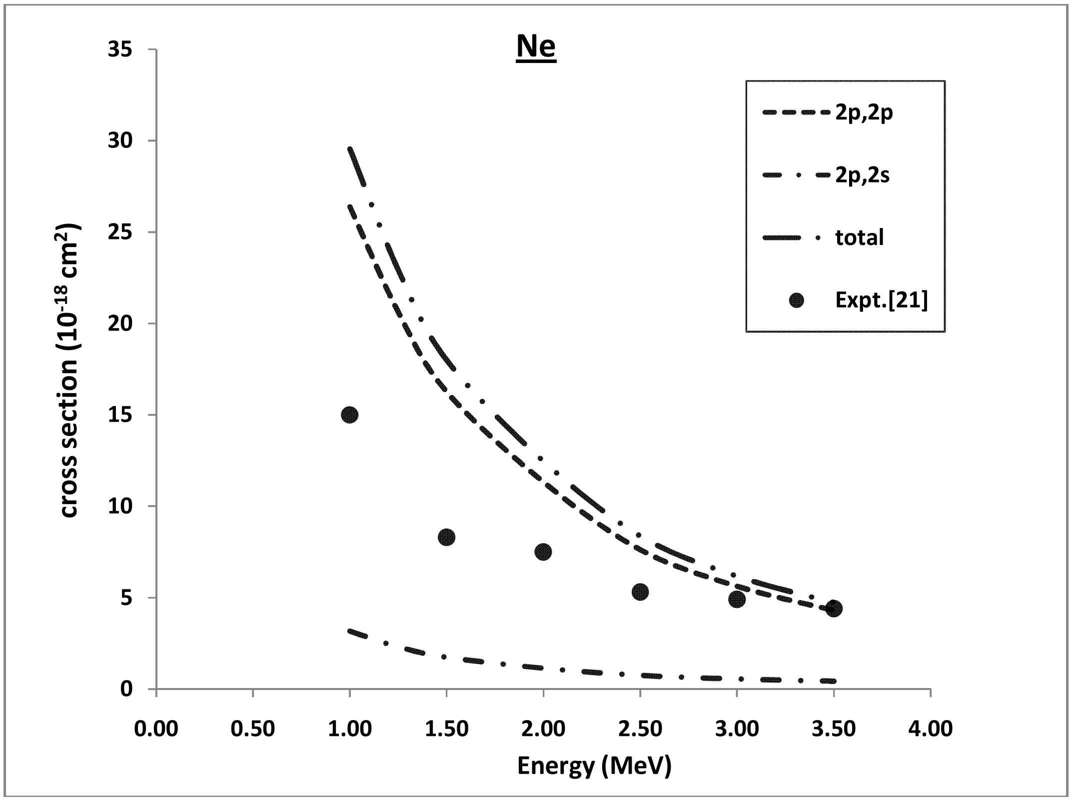

Figure 1: Double ionization of Neon atom by He^+ impact in units of 10^{-18} cm^2

In this case, we have taken the contributions of (2p,2p) and (2p,2s) shells for the calculation of double ionization cross sections. As reported in the paper of Santos [21], theoretical studies of multiple ionization by dressed projectiles are almost in existent and they compared their results of Ne with quantum mechanical calculations of Kirchner et al [22-23]. The comparison shows increasing discrepancies between theory and experiment as the recoil-ion charge state increases. The present cross sections are always within a factor 2 from the experimental results except at energy 1.50 MeV. Moreover, the shapes of the experimental and theoretical calculations curves are in close agreement. At the highest energy say 3.50 MeV the ratio of calculated cross sections to the experimental data is 1.07 and at this energy the magnitudes of both the cross sections are 4.73 x 10^{-18} cm^2 and 4.4 x 10^{-18} cm^2 respectively. Our calculated results throughout overestimates the measured data in the entire energy range, but with the increase of energy both results come close to each other and at the highest energy it is almost similar.

Now we would like to discuss about the double ionization cross sections of Ar. The calculated double ionization cross sections along with the experimental data have been shown in the Table 2 and Figure 2.

Table 2: Double ionization of Argon atom by He^+ impact in units of 10^{-18} cm^2

Energy (MeV)	3p,3p	3p,3s	3p,2p	total	Expt. [21]
1.00	39.81	5.35	0.55	45.71	30
1.25	27.74	3.93	0.53	32.20	
1.50	21.10	3.10	0.53	24.73	21
2.00	13.65	2.08	0.49	16.22	17
s2.50	9.77	1.48	0.44	11.69	11
3.00	7.95	1.18	0.43	9.56	8.5
3.50	5.82	0.84	0.35	7.01	9.1

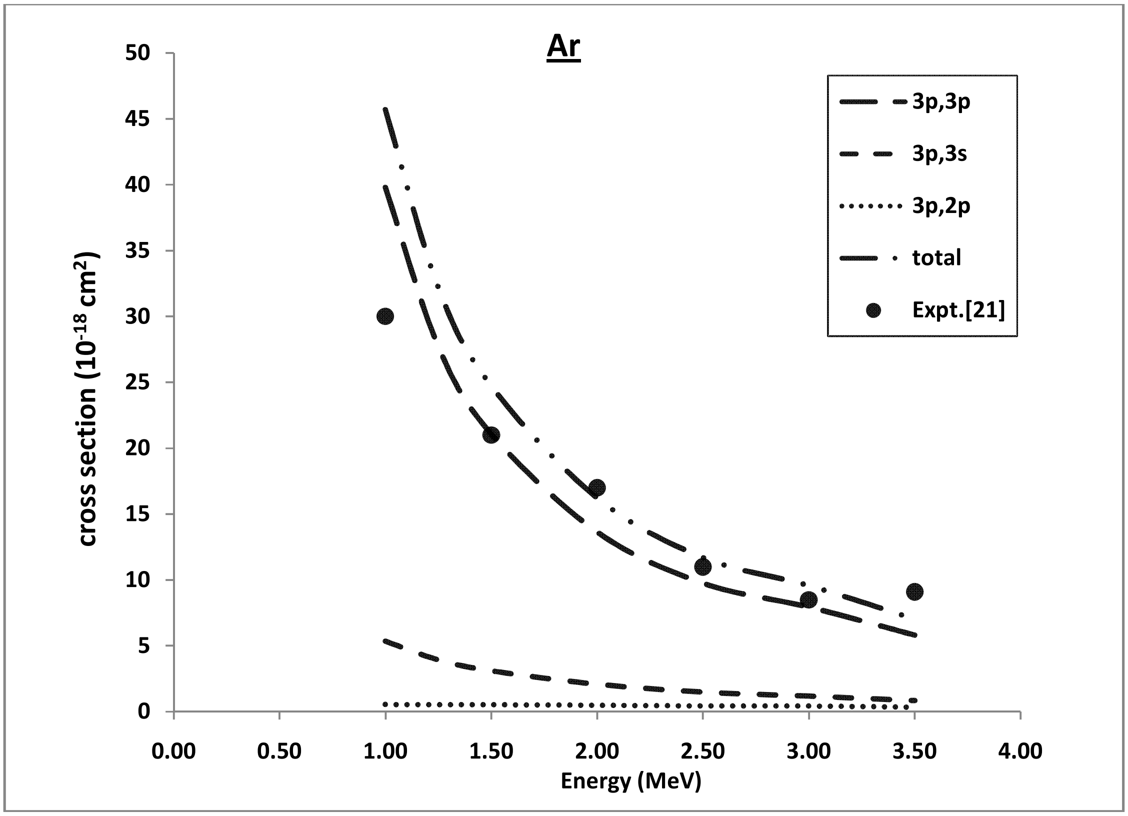

Figure 2: Double ionization of Argon atom by He^+ impact in units of 10^{-18} cm^2

In this calculation we have taken the contributions of (3p,3p), (3p,3s) and (3p,2p). we have compared the calculated results with the experimental measurement of Santos et al [21] in the energy range 1.0 MeV to 3.5 MeV. Our calculated cross sections overestimate the experimental results from 1.0 MeV to 3.0 MeV but experimental result overestimate the cross section at energy 3.5 MeV. The calculated cross sections are within the factor 2 throughout the energy range. At energy 1.0 MeV the ratio of theoretical result to the measured data is 1.52 and at this energy the magnitudes of the calculated result and the experiment are 45.71 x 10^{-18} cm^2 and 30 x 10^{-18} cm^2 respectively. At the energy 2.50 MeV the ratio is 1.06 having the magnitudes 11.69 x 10^{-18} cm^2 and 11 x 10^{-18} cm^2 respectively. At this energy both the results comes very close to each other. Besides this, at the energies 3.00 MeV and 3.50 MeV the ratio of the calculated results to the measured data are 1.12 and 0.77 respectively. From the critical analysis of the results obtained by the present calculation and the experimental data clearly reflects that with the increase of energy both the results comes very close to each other. Hence we find that our calculated cross sections are in good agreement with the experimental data.

Calculated absolute total cross sections for He^+ impact double ionization of Kr from 1.00 MeV to 3.50 Mev are shown in the Table 3 and Figure 3.

Table 3: Double ionization of Krypton atom by He^+ impact in units of 10^{-18} cm^2

Energy (MeV)	4p,4p	4p,3d	4p,4s	4p,3p	total	Expt. [21]
1.00	51.63	8.10	7.39	1.10	68.22	47
1.25	36.87	7.56	5.43	1.07	50.93	
1.50	28.06	7.03	4.16	1.01	40.26	23
2.00	17.84	5.93	2.60	0.86	27.33	23
2.50	12.28	4.96	1.75	0.70	19.69	18
3.00	8.92	4.17	1.25	0.57	14.91	14
3.50	6.73	3.52	0.93	0.47	11.65	12

We have also calculated the cross sections of (4p,4p), (4p,3d), (4p,4s) and (4p,3p) separately. The present results overestimate the experimental data from 1.00 Mev to 3.00 MeV while at energy 3.50 MeV the experimental data slightly overestimate the calculated result. At the energy 3.50 MeV the ratio of calculated cross section to the experimental data is 0.97 having the magnitudes 11.65 x 10^{-18} cm^2 and 12 x 10^{-18} cm^2 respectively. With the increase of the impact energy the calculated cross sections are gradually comes close to the experiment. When we compare our calculated cross sections with the measured data we find that the present calculated results are in excellent agreement with experimental data. Throughout the energy range the calculated cross sections are within the factor 2. The magnitudes of the calculated cross section and the experimental data are 68.22 x 10^{-18} cm^2 and 47 x 10^{-18} cm^2 at impact energy 1.0 MeV having the ratio 1.45 while the magnitudes of both cross sections are 11.65 x $10^{`18}$ cm^2 and 12 x 10^{-18} cm^2 respectively at the impact energy 3.50 MeV. At this energy both the results coalesce to each other. At the impact energy 3.00 MeV the magnitudes of the present cross section and the experimental measurement are 14.91 x 10^{-18} cm^2 and 14 x 10^{-18} cm^2 and its ratio is 1.06. The contributions of different shells, its sum and experimental data are also shown in the Figure and Table separately. This will help the readers to understand the dynamics of the system clearly. At the energy 1.50 MeV the ratio is 1.75 and is the maximum difference observed between the calculated results and the experimental data.

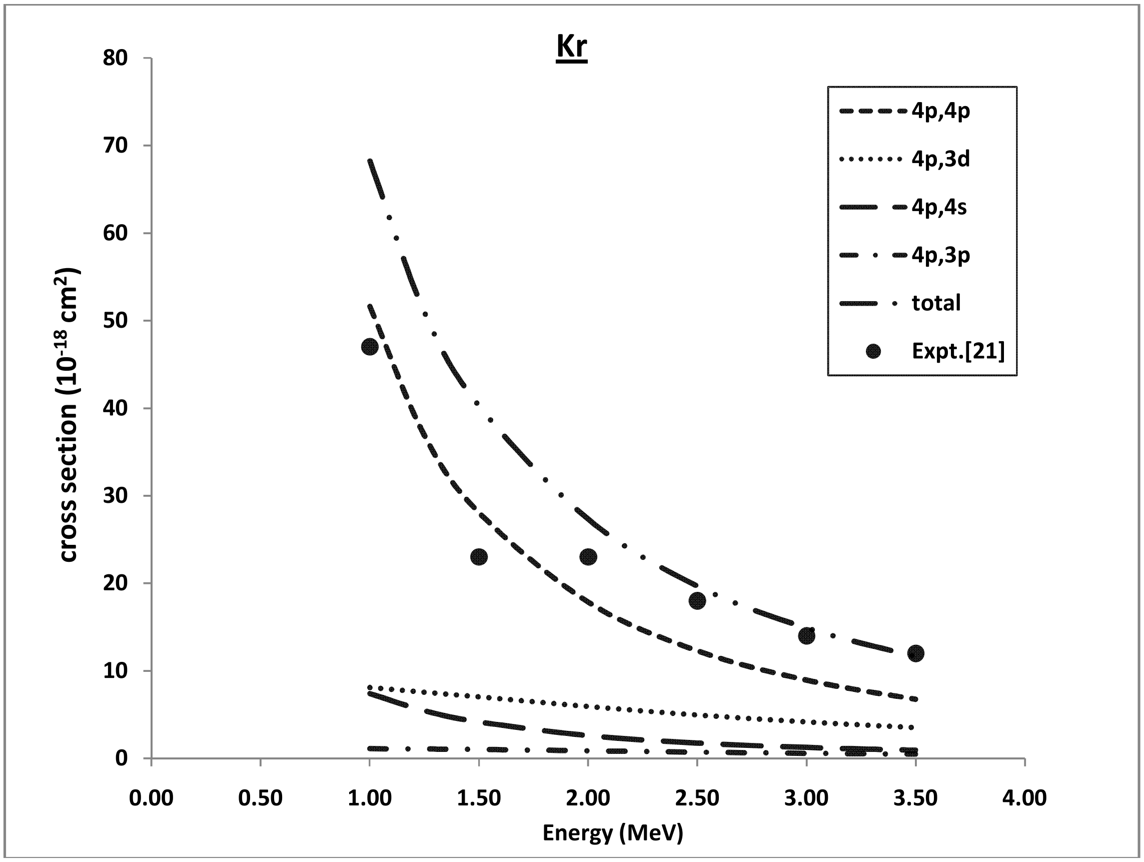

Figure 3: Double ionization of Krypton atom by He$^+$ impact in units of 10^{-18} cm^2

The calculated cross sections for double ionization of Xe from energy range 1.00 MeV to 3.5 MeV along with the experimental data are shown in the Table 4 and Figure 4 respectively.

Table 4: Double ionization of Xenon atom by He$^+$ impact in units of 10^{-18} cm^2

Energy (MeV)	5p,5p	5p,4d	5p,5s	5p,4p	total	Expt [21]
1.00	150.86	31.50	14.81	3.93	201.10	41
1.25	100.14	26.43	10.11	3.41	140.09	
1.50	71.28	22.19	7.32	2.91	103.70	32
2.00	41.68	15.95	4.39	2.11	64.13	27
2.50	27.55	11.81	2.99	1.55	43.90	27
3.00	19.66	8.98	2.19	1.18	32.01	24
3.50	14.73	6.97	1.68	0.92	24.30	20

We have also shown the separate contributions of different shells in the Table 4 and Figure 4. In the case of double ionization cross sections of Xe we have also taken the contributions of (5p, 5p), (5p, 4d), (5p, 5s) and (5p, 4p) shells. The present result differs very much when compared to the experimental data from energy 1.00 MeV to 2.00 MeV and its ratios are 4.90, 3.24 and 2.37 at impact energies 1.00 MeV, 1.50 MeV and 2.00 MeV respectively. From the impact energy 2.50 MeV to 3.50 MeV the calculated results are always within a factor 2. With the increase of the impact energy the present calculated cross sections gradually comes closer to the experimental data. The present theoretical results overestimate the measured data throughout the considered energy range. At the impact energy 1.00 MeV the magnitudes of the calculated result and the experimental data are 201.10 x 10^{-18} cm^2 and 41 x 10^{-18} cm^2 while at the impact energy 3.50 MeV the magnitudes are 24.30 x 10^{-18} cm^2 and 20 x 10^{-18} cm^2 respectively. At the energy 3.50 MeV the ratio of the calculated result to the experimental data is 1.21. The larger value of calculated cross section at low impact energy is the usual trend of Binary Encounter Model. The present model works very well in the case of intermediate and high energy range. Overall microscopic observation of the results shows that the present results are in fairly good agreement with the experimental data.

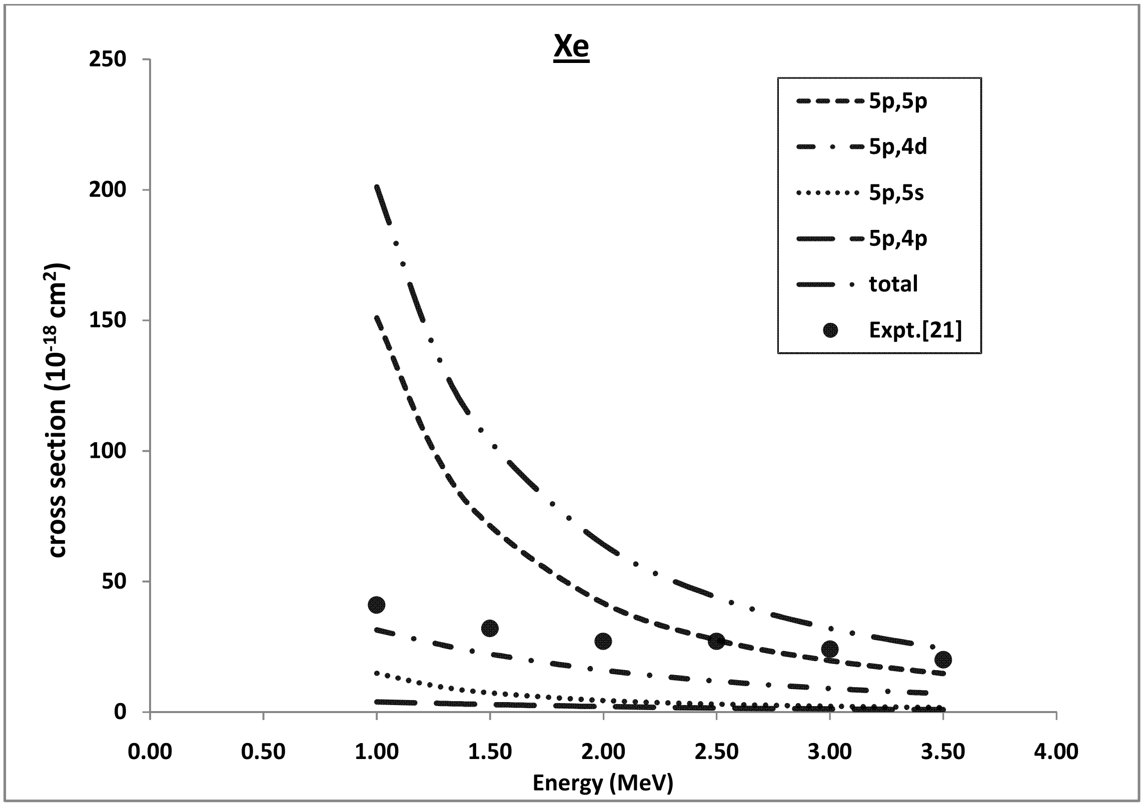

Figure 4: Double ionization of Xenon atom by He$^+$ impact in units of 10^{-18} cm^2

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