

## **Study of proton and alpha particle impact double ionization of Fe**

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### **Abstract**

Theoretical calculations of  $H^+$  and  $He^{2+}$  impact double ionization cross sections for ground state Fe atoms have been performed in the binary encounter approximation (BEA) in the energies region ranging from 80 to 1440 keV/amu for proton impact and from 47 to 360 keV/amu in the case of alpha particle impact. The accurate expression for  $\sigma_{\Delta E}$  (cross section for energy transfer  $\Delta E$ ) and Hartree-Fock velocity distributions for the target electrons have been used throughout the calculations. It has been concluded that the calculated results of  $H^+$  and  $He^{2+}$  impact double ionization cross sections are in good agreement with the experimental data throughout the given energy range.

### **INTRODUCTION**

Among different multiple ionization processes the double ionization is most important. These calculations are considered to be much significance because contribution from different physical process can be separately estimated at various impact energies. The rigorous theoretical calculation of direct double ionization becomes extremely difficult as it needs consideration of four charged particles in the final channel interacting through the long range coulomb potential<sup>1</sup>. Hence, sophisticated calculations of the integrated double ionization cross sections of many electron- atoms by ion impact are not available in the literature.

However, in the past the BEA has been used successfully in the calculation of the charged particle impact single and double ionization cross section for several atoms and ions. Gryzinski reasonably considered two processes in a double BE model to describe direct double ionization<sup>2</sup>. 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. Alternatively, the incident particle may knock out only one target electron and the second electron is removed by the first ejected electron. The idea of above mentioned two step interactions has been supported by number of workers<sup>3, 4</sup>. Roy and Rai modified Gryzinski's theory of electron impact double ionization cross section suitably. In these calculations Hartree-Fock (HF) and hydrogenic velocity distribution were used while considering ejection of the first and second target electrons separately. Later on Jha and Roy used HF velocity distribution while considering the ejection of both electrons of the target in the single and double ionization cross sections of Mg and Ar<sup>5, 6</sup>. The calculated results in BEA are in good agreement with the experimental data. Calculation of double ionization cross sections of titanium-ion by electron impact also shows satisfactory agreement with the experimental observation<sup>6</sup>.

In the case of heavy charged particle impact the BE calculations of double ionization cross section of atoms are very few in the literature. Kumar and Roy<sup>7</sup> pointed out some error in the Gryzinski's theory for calculation of the above mentioned processes and modified the mathematical frame work suitably incorporating the necessary corrections using the accurate expression of  $\sigma_{\Delta E}$  as given by Vriens<sup>8</sup>, they calculated proton impact double ionization cross section of He, Ne, Ar, and Kr, which were found to be satisfactory agreement with the experimental observation<sup>7, 9</sup>. In these calculations hydrogenic and HF velocity distribution were used for considering the ejection of the first and second target electrons respectively. From comparison of two distribution functions they have concluded that the use of HF velocity distribution for the ejection of both electrons in calculations of direct double ionization would lead to better agreement with the experimental data. Singh et al. has calculated proton and alpha particle impact single and double ionization cross sections of Mg atom and found reasonably good agreement with the experiment<sup>10</sup>. Keeping the above mentioned facts in view, we have considered it worthwhile to carry out calculations of H<sup>+</sup> and He<sup>2+</sup> double ionization cross sections for iron atoms in BE using HF velocity distribution for both the ejected electrons. This work will encourage us to critically analyze direct double ionization cross sections and to verify the contribution 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 the 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 binding energy of the atomic electron<sup>11</sup>. Here we have assumed  $Z^2$  dependence also in calculation of direct double ionization cross sections in the present double binary encounter model, justifications of which will be after the presentation of the mathematical expression.

In the present work we have used the accurate expression of  $\sigma_{\Delta E}$  (cross section for energy transfer  $\Delta E$ ) as given by Vriens for heavy charged particles incident on atoms<sup>8</sup>. Following Catlow and McDowell we have introduced two dimensionless variables  $s$  and  $t$  defined by  $s^2 = v_1^2 / v_0^2$  and  $t^2 = v_2^2 / v_0^2$ , where  $v_1$  and  $v_2$  are the velocities in atomic units of the incident particle and the target electron respectively and  $u = v_0^2$  is the ionization potential of the target in rydbergs<sup>11</sup>. All other energies involved are also expressed in rydbergs. In terms of these variables, the expressions of ionization cross section due to a projectile of unit charge

for a particular incident energy and a particular velocity of bound electron are given by (see Kumar and Roy<sup>7</sup>)

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

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

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

(1)

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.

Heavy charged particle impact double ionization cross section  $Q_D^{ii}$  is given by

$$Q_D^{ii} = Q_{sc}^{ii} + Q_{ej}^{ii} \quad (2)$$

In accordance of the idea given by Gryzinski<sup>2</sup> in double binary encounter model, these cross sections involving integrals over energy transfer are given by

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

(3)

and

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

(4)

where  $n_e$  is number of electrons in a shell and  $f(t)$  is the momentum distribution function of the target electrons and defined as

$$f(t) = 4\pi t^2 \rho_{nl}(u_i^{1/2} t) u_i$$

where

$$\rho_{nl} = \frac{1}{2l+1} \sum_{m=-l}^{m=l} |\psi_{nlm}(x)|^2$$

and

$$\psi_{nlm}(x) = \frac{1}{(2\pi)^{3/2}} \int \Phi_{nlm}(r) e^{ix \cdot r} dr \text{ is the Fourier transform of the one electron}$$

orbit and

$$\Phi_{nlm}(r) = N_{nl} R_{nl}(r) Y_{lm}(\Omega)$$

where  $R_{nl}(r)$  is the analytical Hartree-Fock radial function which has been taken from Roothaan et al.<sup>12</sup>.

The symbols used in the above expressions have been defined by Gryzinski<sup>2</sup>. Here  $\Delta E$  and  $\Delta E'$  stand for energy transfer during the first and the second collisions respectively and  $\bar{r}$

denotes the mean distance between the electrons in the shell 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 the case of a projectile of unit charge is given by (see Kumar and Roy<sup>7</sup>)

$$\sigma_{\Delta E} d(\Delta E) = \begin{cases} Ad(\Delta E); & \Delta E \leq 4su(s-t) \\ Bd(\Delta E); & 4su(s-t) \leq \Delta E \leq 4su(s+t) \\ 0; & \Delta E > 4su(s+t) \end{cases} \quad (5)$$

where

$$A = \frac{4}{s^2 u} \left( \frac{1}{(\Delta E)^2} + \frac{4t^2 u}{3(\Delta E)^3} \right) \quad \text{and} \quad B = \frac{2}{3t(\Delta E)^3} \left( 8s - \frac{[(\Delta E + t^2 u)^{1/2} - tu^{1/2}]^3}{s^2 u^{3/2}} \right) \quad (6)$$

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 electrons are given below.

$$Q_{sc}^{ii} = \frac{n_e(n_e-1)Z^2}{4\pi\bar{r}^2} \times \left( \int_{t=0}^{s-\frac{1}{4s}} \left\{ \int_{u_i}^{4su_i(s-t)} A\alpha d(\Delta E) + \int_{4su_i(s-t)}^{4su_i(s+t)} B\alpha d(\Delta E) \right\} f(t)u_i^{1/2} dt + \int_{t=s-\frac{1}{4s}}^{\infty} \int_{u_i}^{4su_i(s+t)} B\alpha f(t)u_i^{1/2} d(\Delta E) dt \right) (\pi a_0^2) \quad (7)$$

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

$$Q_{sc}^{ii} = \frac{n_e(n_e-1)Z^2}{4\pi\bar{r}^2} \times \left( \int_{t=\frac{1}{4s}-s}^{\infty} \int_{u_i}^{4su_i(s+t)} B\alpha f(t)u_i^{1/2} d(\Delta E) dt \right) (\pi a_0^2) \quad \text{when } (s-1/4s) \text{ is negative} \quad (8)$$

$$\text{In the above expressions} \quad \alpha = \int_0^{\infty} Q_i(s', t) f'(t) u_{ii}^{1/2} dt (\pi a_0^2)$$

(9)

Here  $s'$  is given by

$$s'^2 = \begin{cases} \frac{E_q - \Delta E}{1836u_{ii}} \text{ for } H^+ \text{ impact} \\ \frac{E_q - \Delta E}{7344u_{ii}} \text{ for } He^{2+} \text{ impact} \end{cases}$$

(10)

Similarly equations for ejected part are

$$Q_{ej}^{ii} = \frac{n_e(n_e-1)Z^2}{4\pi r^2} \times \left( \int_{t=0}^{s-(1+\frac{u_{ii}}{u_i})/4s} \left\{ \int_{u_i+u_{ii}}^{4su_i(s-t)} A\alpha' d(\Delta E) + \int_{4su_i(s-t)}^{4su_i(s+t)} B\alpha' d(\Delta E) \right\} f(t)u_i^{1/2} dt + \int_{t=s-(1+\frac{u_{ii}}{u_i})/4s}^{\infty} \int_{u_i+u_{ii}}^{4su_i(s+t)} B\alpha' f(t)u_i^{1/2} d(\Delta E) dt \right) (\pi a_0^2) \quad (11)$$

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

$$Q_{ej}^{ii} = \frac{n_e(n_e-1)Z^2}{4\pi r^2} \times \left( \int_{t=(1+\frac{u_{ii}}{u_i})/4s-s}^{\infty} \int_{u_i+u_{ii}}^{4su_i(s+t)} B\alpha' f(t)u_i^{1/2} d(\Delta E) dt \right) (\pi a_0^2) \quad (12)$$

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

$$\alpha' = \int_0^{\infty} q_i(s', t) f'(t) u_{ii}^{1/2} dt (\pi a_0^2) \quad (13)$$

Here  $q_i(s', t)$  is the expression for electron impact ionization cross section of atoms (see Jha and Roy<sup>5</sup>) and  $s'$  is given by  $s'^2 = \frac{\Delta E - u_i}{u_{ii}}$  for both  $H^+$  and  $He^{2+}$  impact.

Now we discuss the  $Z^2$  dependence of the expression of  $Q_{sc}^{ii}$  which denote a process in which the projectile knock 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<sup>3</sup> and 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 contribution of  $Q_{ej}^{ii}$  are found to be much smaller than those of  $Q_{sc}^{ii}$  (see Kumar and Roy<sup>7,9</sup>). In case of proton impact  $Z = 1$  and therefore  $Z^4$  scaling for  $Q_{sc}^{ii}$  become essentially the same as  $Z^2$  scaling and good agreement of calculated results with the experiment is achieved. However, in the case of alpha particle impact calculation involves  $Z = 2$  and a  $Z^4$  scaling of  $Q_{sc}^{ii}$  lead to much dominant contribution of the process adversely affecting the results. Hence the correspondence of the processes represented by  $Q_{ej}^{ii}$  and the  $Q_{sc}^{ii}$  to the first and the second Born approximation does not appear to be suitable. In this contest the experimental results of  $H^+$  and  $He^{2+}$  impact pure double ionization cross sections are noteworthy.

The integral 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 distributions corresponding to the first and the second ejected electron respectively. These have been constructed from HF radial wave functions (see Catlow and McDowell<sup>13</sup>, Jha and Roy<sup>5</sup>). We have considered total cross section for heavy charged particle impact direct double ionization of Fe as given by

$$Q_D^{ii} = Q_D^{ii}(4s,4s) + Q_D^{ii}(4s,3d) + Q_D^{ii}(4s,3p) \quad (14)$$

The factor  $\frac{n_e(n_e-1)}{4\pi^2}$  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  $n_{e1} \times n_{e2}$  where these two stand for number of electrons in the shells under consideration. The binding energies of the shells of Cu, the expectation values of the shell radii and HF radial wave functions have been taken from the data reported by Clementi and Roetti<sup>14</sup>.

## RESULT AND DISCUSSION

### Fe+ H<sup>+</sup> impact Double ionization

In the case of H<sup>+</sup> impact double ionization of Fe we have considered contribution of (4s,4s), (4s,3d) and (4s,3p) shells. In this case we have compared our calculated results with the experimental data of Patton et al.<sup>15</sup>. We have calculated the cross sections from energy range 93 to 1440 keV/amu. In our calculated results we found that the contributions of (4s,4s), (4s,3d) and (4s,3p) having magnitudes  $2.16 \times 10^{-17} \text{ cm}^2$ ,  $14.66 \times 10^{-17} \text{ cm}^2$  and  $1.49 \times 10^{-17} \text{ cm}^2$  at impact energies 93 keV/amu. At this energy range the experimental results having magnitude  $5.60 \times 10^{-17} \text{ cm}^2$  while the calculated results having magnitude  $18.31 \times 10^{-17} \text{ cm}^2$  respectively. The ratio of the theoretical calculation to the experimental data is 3.26 at the lowest energy. Both the experimental data and the calculated cross sections is highest at the lowest energy while it gradually decreases with the increase of energies considered. A low energy the theoretical results dominates to the experimental data while with the increase of energy the calculated results decreases rapidly as compared to the experimental measurements. With the increase of energy both the results are coming closer to each other and at the energy 720 keV/amu it is almost similar. At this energy the calculated result is of magnitude  $1.84 \times 10^{-17} \text{ cm}^2$  and the experimental data is of the magnitude  $1.81 \times 10^{-17} \text{ cm}^2$ . Beyond this energy the magnitude of the calculated results gradually decreases as compared to the magnitude of experimental data. At the highest energy 1440 keV/amu the magnitude of calculated result is  $0.61 \times 10^{-17} \text{ cm}^2$  while the experimental data is of magnitude  $1.06 \times 10^{-17} \text{ cm}^2$ . At this energy the ratio of calculated results to the experimental data is 0.57. From the energy range 93 keV/amu to 250 keV /amu the results are beyond the factor of 2. But with the increase of energy both the results are coming close to each other. From the energy range 360 keV/amu to 1440 keV/amu the results are within the factor of 2. From the close inspection the magnitude of experimental measurement decreases slowly while the magnitudes of theoretical results are decreasing very rapidly i.e. the ratio of the lowest to highest energies cross sections in the case of theoretical to experimental are more than thirty times and more than five times respectively. The overestimation of the calculated results at low energy range is the usual feature of our calculation (i.e. following BEA) The ratio of the calculated cross section to the experimental measurements are 2.0, 1.44, 1.01, 0.88, 0.64, 0.57 at impact energies 360, 500, 720, 850, 1200, and 1440 keV respectively. At the energies 360, 500, 720, 850, 1200 and 1440 keV/amu the magnitudes of the calculated results are  $5.01 \times 10^{-17} \text{ cm}^2$ ,  $3.17 \times 10^{-17} \text{ cm}^2$ ,  $1.84 \times 10^{-17} \text{ cm}^2$ ,  $1.4 \times 10^{-17} \text{ cm}^2$ ,  $0.80 \times 10^{-17} \text{ cm}^2$  and  $0.61 \times 10^{-17} \text{ cm}^2$  while the experimental results having magnitudes  $2.5 \times 10^{-17} \text{ cm}^2$ ,  $2.2 \times 10^{-17} \text{ cm}^2$ ,  $1.81 \times 10^{-17} \text{ cm}^2$ ,  $1.6 \times 10^{-17} \text{ cm}^2$ ,  $1.24 \times 10^{-17} \text{ cm}^2$  and  $1.06 \times 10^{-17} \text{ cm}^2$  respectively. From the discussion given above it clearly indicates that

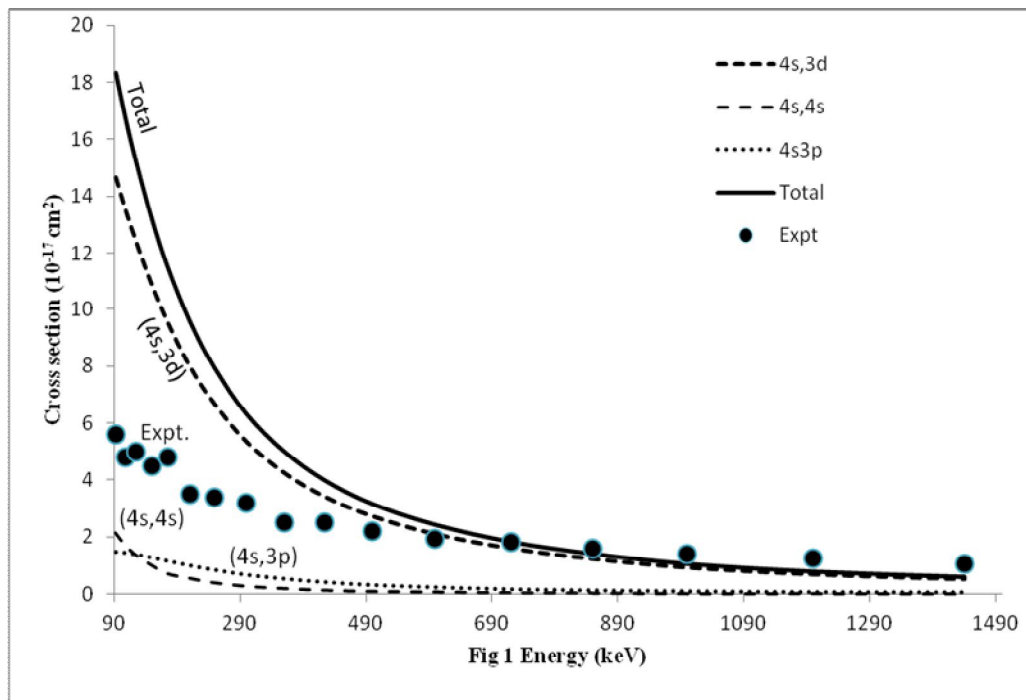
with the increase of energy the results are coming close to each other and are within the factor of 2 and supposed to be in good agreement with the experimental results

**Table 1:** Proton impact double ionization cross sections of Fe in unit of  $10^{-17} \text{ cm}^2$

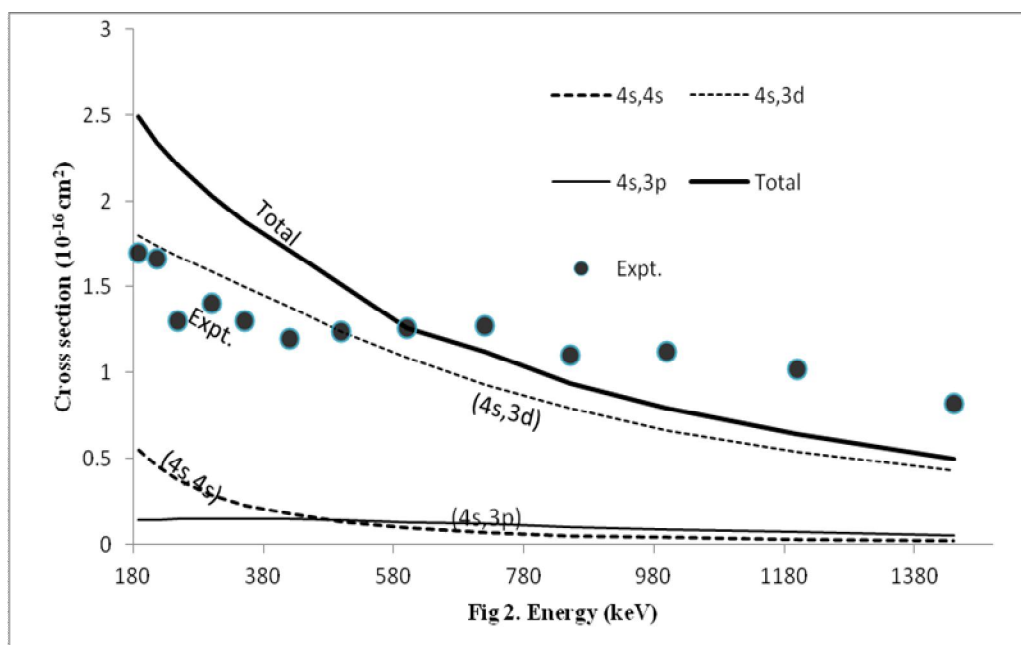
Energy(keV/amu)	Contribution of (4s,3d)	Contribution of (4s,4s)	Contribution of (4s,3p)	Total	Expt.[15]
93	14.66	2.16	1.49	18.31	5.6
108	13.55	1.74	1.46	16.75	4.8
125	12.42	1.39	1.40	15.21	5
150	10.88	1.03	1.29	13.2	4.5
175	9.55	0.73	1.17	11.51	4.8
210	8.01	0.57	1.01	9.59	3.5
250	6.66	0.41	0.85	7.92	3.4
300	5.36	0.29	0.68	6.33	3.2
360	4.26	0.21	0.54	5.01	2.5
425	3.42	0.15	0.42	3.99	2.5
500	2.74	0.11	0.32	3.17	2.2
600	2.11	0.09	0.24	2.44	1.92
720	1.61	0.06	0.17	1.84	1.81
850	1.24	0.05	0.13	1.42	1.6
1000	0.94	0.03	0.10	1.07	1.38
1200	0.70	0.03	0.07	0.80	1.24
1440	0.53	0.02	0.06	0.61	1.06

**Table 2:** Alpha particle impact double ionization cross section of Fe in unit of  $10^{-16} \text{ cm}^2$

E(keV/amu)	Contribution of (4s,4s)	Contribution of (4s,3d)	Contribution of (4s,3p)	Total	Expt.[15]
47	0.55	1.80	0.14	2.49	1.7
54	0.46	1.74	0.14	2.34	1.66
62	0.38	1.68	0.15	2.21	1.3
75	0.29	1.59	0.15	2.03	1.4
88	0.23	1.50	0.15	1.88	1.3
105	0.18	1.38	0.15	1.71	1.2
125	0.13	1.24	0.14	1.51	1.24
150	0.10	1.09	0.13	1.26	1.26
180	0.07	0.93	0.12	1.12	1.27
213	0.05	0.79	0.10	0.94	1.1
250	0.04	0.66	0.09	0.79	1.12
300	0.03	0.54	0.07	0.64	1.02
360	0.02	0.43	0.05	0.50	0.82



**Figure 1:** Proton impact double ionization cross section of Fe:  
The (4s,4s), (4s,3d) and (4s, 3p) stand for partial contribution to the proton impact direct double ionization and Total stands for the total theoretical double ionization cross section of Fe and experimental data as Expt.[15]



**Figure 2:** Alpha particle impact double ionization cross section of Fe:



The (4s,4s), (4s,3d) and (4s, 3p) stand for partial contribution to the electron impact direct double ionization of Fe and Total stands for the total theoretical double ionization cross section of Fe and experimental data as Expt.[15]

### He<sup>2+</sup> impact Double Ionization of Fe

In the calculation of He<sup>2+</sup> impact double ionization of Fe we have calculated the cross sections from the energy range 47 keV/amu to 360 keV/amu and compared with the experimental data of Patton et al.<sup>15</sup>. In this calculation we have taken the contribution of (4s,4s), (4s,3d) and (4s,3p) shells only. In this calculation from the lower energy range to 125 keV/amu the calculated cross sections overestimate the measured data. Beyond this energy the calculated cross sections underestimates the contributions of the experimental cross sections up to the highest energy which we have considered, except at 150 keV/amu. The overestimation of the calculated cross sections at low energy range is the usual trend of BEA. The ratio of the calculated cross sections to the experimental measured data is almost within the factor of 2 and it becomes identical at energy of 150 keV/amu. At the lower energies 47 keV/amu, 54 keV/amu, 75 keV/amu the ratios of calculated to experimental cross sections are 1.46, 1.40 and 1.45 respectively. But the increase of the energies at 150, 213, 300 and 360KeV/amu the ratio of the calculated cross sections to the experimental data are 1.0, 0.85, 0.62 and 0.60 respectively. The experimental cross sections at lowest energy 47 keV/amu is of magnitude  $1.7 \times 10^{-16} \text{ cm}^2$  while at the highest energy of 360 keV/amu it becomes  $0.82 \times 10^{-16} \text{ cm}^2$ . The ratio of the lowest cross section to the highest cross section is almost doubled while the ratio of the lowest calculated cross section to the highest cross section is all most five times greater which indicates that the fall in magnitude of the experimental cross sections decrease slowly while the calculated cross sections fall rapidly with the increase of energy as compared to the experimental findings.

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