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Electron impact single and double ionization of Fe atom: PACS. 34.80 Dp Atomic excitation and ionization by electron impact

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Abstract:

Theoretical calculations of electron impact single and double ionization cross sections for ground state Fe atoms have been performed in the binary encounter approximation(BEA) in the energy region ranging from threshold to 1250 eV and 1200eV respectively. The accurate expression for $\sigma_{\Delta E}$ (cross section for energy transfer ΔE) including exchange and interference as given by Vriens and Hartree – Fock velocity distributions for the target electrons have been used throughout the calculations.

INTRODUCTION

Various processes can contribute to electron impact double ionization of atoms and ions depending on the incident electron energy and on the structure of parent and intermediate atomic states. For direct ejection of two outer shell electrons, two different types of mechanism are identified: shake off and two state mechanism. In addition, many indirect double ionization processes are associated with the formation of auto ionizing states following inner shells ionization or excitation.

In the case of direct double ionization (DDI) via shake off the incident electron interacts with a bound electron and ejects it with outer bound electrons being left in the state which is not an Eigen state of the residual ion. In the subsequent relaxation process there is finite probability of a second ionization. Electron impact double ionization of atoms and ions is a four particle (one ion and three electrons) problem. In the final channel, these four charged particles interacts with each other via long range Coulomb potential, making the correct treatment of this many body problem extremely difficult. For this reason, it is still impossible to carry out exact calculations for these processes, both full theoretical calculation and detail experimental investigations remain scare. On the other hand, the most detailed description of the process is given by means of full differential cross sections allowing the analysis of angular and energy distributions for each one of the ejected or scattered electrons. On the other hand the total double ionization cross sections describe the global importance of non charge state. Electron impact ionization of ions is a fundamental process in any discharge or plasma so that measurement of absolute total ionization cross section of various ions have long been performed , including the case of double ionization where a number of reports are available.

Electron impact ionization of atoms and ions is one of the most fundamental collision processes in atomic and molecular physics. Knowledge of ionization cross section for these processes finds wide application in plasma kinematics problems, mass spectroscopy, gas laser, upper atmospheric physics and astrophysics. Ionization rate of various atomic species found in astronomical plasma are also a great interest. From an applied view point multiple ionization process are important in moderate and high temperature plasma in all gaseous environment with an abundant energetic electrons [1].

Experimental investigation of ionization cross section for metals lead to several difficulties and have been carried out by only very few experimental groups for limited number of elements. Accurate experimental measurement of multiple ionization of iron by electron impact have been carried out by Shah et al.[2] using pulse cross beam technique incorporating time - of –flight spectroscopy of the collision products to study the electron impact ionization of ground state Fe atom within the energy range from respective thresholds to 1250eV . Experimental data obtained by Shah et al. [2] would not be compared with previous theoretical calculation of double ionization cross section due to non availability of the data in the literature.

Individual cross sections have been obtained by normalization to lower energy values of σ_2 . Previously measured by Freund et al. [3] using a fast crossed beam experiment analysis was complicated by the presence of metastable atom in Fe-beam. Measured cross sections exhibit evidence of contribution from inner shell electrons. The high energy values of single ionizations are in good agreement with theoretical predictions based on the first Born approximation.

Sophisticated theoretical calculations of single and double ionization cross section of Fe are not available in the literature. Rigorous theoretical calculation of double ionization cross section becomes very complicated as it is related with the consideration of the four charged particles in the final channel interacting through the long range Coulomb potential [4]. Quantal calculation of the integrated double ionization cross sections of atoms/ions by electron impact have not been reported so far. Recently Belenger et al. [5] have reported a semi empirical formula for evaluation of double ionization cross section of neutral atoms, and positive and negative ions by electron impact and also presented results for Cu-target. In this approach the shape of the cross section is described by analytical expression and approximation -parameters are estimated by fitting the model cross sections to reliable experimental data. Besides this, similar methods have been reported by Fisher el al. [6] and Deutsch et al. [1]

A few attempts have been made to calculate electron impact double ionization cross section for light target e.g. H^+, He and Li^+ is the Born approximation (see Tweed [7, 8] and Mc Guire [9]. In a promising approach the time dependent close coupling method was used by Pindzola et al.[10] in the calculation of electron impact double ionization of cross section of He. Afterward Pindzola and his coworkers carried out calculations of electron impact double ionization cross section of Mg [11] , Be [12] and B^+ ion [13] using a non perturbative time dependent close coupling method. However, such calculations are restricted so far to essentially two electrons system. Using classical binary encounter approximation (BEA), Gryzinski and Kun [14] have derived general analytical expression for electron impact double ionization cross section of atoms with atomic number Z ≥ 20 and *s* or *d* outer shell with two electrons. They have compared their calculations only with experimental data for Ca, Sr, Ba and Hg atoms and found satisfactory agreement. Although, this model is consistent and convenient but it treats processes of double ionization in statistical ways. However, this model is not applicable in case of Fe. Here we would like to mention that the wave functions representing the bound electrons are characteristic of the target atoms but there is no consideration of wave function in above mentioned calculation.

Keeping in view the above mentioned fact, we have used the symmetrical collision model of Vriens including exchange and interference in the present work and used Hartree-Fock velocity distribution for the target electrons. We have also taken into consideration of inner shells. In the past, the BE approximation has been found successful in the calculation of electron impact single and double ionization of atoms [15-17].

THEORETICAL METHODS

We have used the accurate expression for $\sigma_{\Delta E}$ including exchange and interference as given by Vriens [18] for calculating electron impact single ionization cross sections. The expression used in the calculation has been discussed in detail by Roy and Rai [19]. A brief presentation of the expression in final form used in the calculations is given below. Using dimension less variables introduced by Catlow and McDowell [20], the expression for cross section for particular incident energy and a particular velocity of the bound electron can be written as in the form

$$Q^i(s,t) = \frac{4}{(s^2+t^2+1)u^2}\left[\frac{s^2-1}{s^2} + \frac{2t^2}{3}\left(\frac{s^4-1}{s^4}\right) - \frac{\phi \ln s^2}{(s^2+1)}\right](\pi a_0^2)$$

$$\text{where} \qquad \phi = \cos\left\{\left(\frac{1}{s^2u+u}\right)^{1/2} \ln s^2\right\} \tag{1}$$

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. Thus the expression for electron impact single ionization cross section for a particular shell of the target is given by

$$Q^i(s) = n_e \int_0^\infty Q^i(s,t) f(t) u^{1/2} dt \tag{2}$$

The method of calculating electron impact double ionization cross section of atoms in double binary encounter model has been discussed in detail in earlier publications [21, 22, 15]. However, it is desirable to give a brief discussion of the expression which has been used in present calculations. Electron impact double ionization cross sections including contribution from Auger emission can be written as

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

where Q_D^{ii} denotes the contribution from direct ejection of two electron and Q_A^{ii} that from Auger emission. The expressions for cross sections corresponding to the two processes of the double binary encounter model leading to direct double ionization are given by (see Jha and Roy [15]).

$$Q_{SC}^{ii} = \frac{n_e(n_e-1)}{4\pi\bar{r}^2}\int_{t=0}^{\infty}\int_{U_i}^{E_q-U_{ii}}\sigma_{\Delta E}\left[\int_{t=0}^{\infty}\int_{U_{ii}}^{E_q-\Delta E}\sigma_{\Delta E^{`}} f(t)U_{ii}^{1/2}d(\Delta E^{i})dt\right]\times f(t)U_i^{1/2}d(\Delta E)dt\times 8.797\times10^{-17}(\pi a_0^2) \tag{4}$$

and

$$Q_{ej}^{ii} = \frac{n_e(n_e-1)}{4\pi\bar{r}^2}\int_{t=0}^{\infty}\int_{U_i+U_{ii}}^{E_q}\sigma_{\Delta E}\times\left[\int_{t=0}^{\infty}\int_{U_{ii}}^{\Delta E-U_i}\sigma_{\Delta E^{`}} f(t)U_{ii}^{1/2}d(\Delta E^{i})dt\right]\times f(t)U_i^{1/2}d(\Delta E)dt\times 8.797\times10^{-17}(\pi a_0^2) \tag{5}$$

In the case of single ionization, we have used the accurate expression for $\sigma_{\Delta E}$ as given by Vriens [18]in the above expression also. Using dimensionless variables introduced by Catlow and McDowell [20] $\sigma_{\Delta E}$ is given by (see Kumar and Roy [23])

$$\sigma_{\Delta E} = \frac{2}{(s^2+t^2+1)u}\left[\left(\frac{1}{\Delta E^2}+\frac{4t^2u}{3\Delta E^3}\right)+\left(\frac{1}{(s^2u+u-\Delta E)^2}+\frac{4t^2u}{3(s^2u+u-\Delta E)^3}\right)-\frac{\phi}{\Delta E(s^2u+u-\Delta E)}\right] \tag{6}$$

where $\phi = \cos\left\{\left(\frac{1}{s^2u+u}\right)^{1/2}\ln s^2\right\}$

Due to indistinguishability of electron in the symmetrical model of Vriens the cross sections corresponding to the two process are exactly equal at all incident energies (see Kumar and Roy [23]) and hence in order to obtain the direct double ionization cross section, either of the cross sections should be multiplied by two. In equation (4) u and s^2 have been replaced by U_i and E_q/U_i in the expression for $\sigma_{\Delta E}$ and by U_{ii} and $(E_q-\Delta E)/U_{ii}$ in the case of $\sigma_{\Delta E^{`}}$. The only difference in the equation (5) is that $s^2$ assumes the value $(\Delta E-U_i)/U_{ii}$ in the expression for $\sigma_{\Delta E^{`}}$.The function $f(t)$ appearing in equations (2), (4) and (5) is the momentum distribution function (see Catlow and McDowell [20]) and Jha and Roy[15].In case of double ionization $f(t)$ has been constructed replacing u by U_i and U_{ii} for ejection of first and the second electron respectively. In order to obtain Q_A^{ii} (contribution to the double ionization from Auger emission), the single ionization cross section should be multiplied by Auger yield of the shell under consideration.

We have considered total cross section for electron impact direct double ionization of Fe as given by

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

where $Q_D^{ii}(4s,3d)$ stands for the double ionization cross section corresponding to one electron ejected from 4s shell and the other from the 3d shell. The factor $n_e(n_e-1)/4\pi\bar{r}^2$ has been suitably modified for considering the modes of ionization in which the electrons are ejected from different shells. $n_e(n_e-1)$ has been replaced by $n_e1\times n_e2$ where these two stand for number of the electrons in the shell under consideration. In order to obtain the value of $\bar{r}$, the atomic radius has been replaced by the mean of the expectation value of radii of the shell (see Jha and Roy[15]). For binding energies we have used the magnitude of orbital energies of the shells of Fe given by Climenti and Roetti [24] and Hartree-Fock radial wave functions have been used to construct momentum distribution functions for the target electron in the present calculation. The expectation values of radii reported by Desclaux [25] have been used as shell radii.

RESULTS AND DISCUSSION

Electron Impact Single Ionization cross sections of Fe

We have calculated the electron impact single ionization cross sections of Fe from 8.1 eV to 1250 eV impact energies. Such calculations over a wide energy range with an experiment [2] have been reported in Table1 and Figure1. In this calculation we have calculated single ionization cross sections of 4s, 3d and 3p shells. The remaining inner shells contributions are almost negligible and hence contribution of inner shells has not taken into account. Near the threshold the magnitude of the calculated cross sections are greater in compared to the experimental results which is a usual feature of this model. The present model is valid for high energy ranges. Our calculated results come closer to the experimental data with increase of impact energy from 8.6 eV to 1250 eV. The calculated peak has magnitude 6.29 x 10^{-16} cm^2 at 32.5 eV while the peak measured by M B Shah et al. is 4.08 x 10^{-16} cm^2 at 35 eV. The ratio of the calculated peak to the experimental peak is 1.54.The magnitude of the calculated peak is shifted slightly towards lower energy side compared to the experimental peak. The ratio of calculated cross sections to the experimental cross sections becomes greater than 2 above impact energy 8.8 eV. One of the important and noticeable thing is that both calculated cross section 0.87 x 10^{-16} cm^2 and experimental cross section 0.53 x 10^{-16} cm^2 respectively have minimum value at same impact energy 1250 eV. From the close inspection of the curves it is found that the major contributions to the calculated cross sections comes from 4s and 3d sub shells while the contribution of 3p shell is very small. In the calculated results it is found that the contributions of *4s* shell falls sharply with the increase of energy up to 275 eV while the contribution of 3d shells fall smoothly with the increase of energy up to 170 eV. At the impact energy 110 eV the magnitudes of 3d shells and 4s shells are almost similar.

Table 1: Electron impact single ionization cross section of Fe in the unit of 10^{-16} cm^2

E(eV)	Contribution of 4s	Contribution of 3d	Contribution of 3p	Total	Expt[2]
8.10	1.26			1.26	0.33
8.30	1.47			1.47	0.44
8.60	1.76			1.76	0.70
8.80	1.93			1.93	1.08
9.40	2.38			2.38	1.34
9.90	2.70			2.70	1.54
10.40	2.96			2.96	1.96
11	3.23			2.23	2.18
11.60	3.46			3.46	2.22
12	3.59			3.59	2.39
12.60	3.76			3.76	2.46
13	3.85			3.55	2.50
14	4.05			4.05	2.83
15	4.20			4.20	2.94
16.10	4.32			4.32	3.15
16.80	4.38			4.38	3.42
19	4.48	0.45		4.93	3.65
21	4.50	0.93		5.43	3.91

23	4.49	1.29		5.78	3.95
26	4.42	1.67		6.09	4.01
29	4.30	1.92		6.22	3.80
30	4.27	1.98		6.25	3.92
32.50	4.17	2.12		6.29	3.98
35	4.06	2.21		6.27	4.08
42	3.76	2.37		6.13	3.77
47	3.56	2.42		5.98	3.76
52	3.37	2.45		5.82	3.58
60	3.11	2.45		5.56	3.42
70	2.83	2.42		5.25	3.29
80	2.59	2.37	0.02	4.98	2.93
85	2.49	2.34	0.05	4.88	2.93
90	2.39	2.31	0.07	4.77	2.87
95	2.30	2.28	0.08	4.66	2.83
100	2.22	2.25	0.09	4.56	2.60
110	2.07	2.19	0.12	4.38	2.46
120	1.94	2.13	0.13	4.20	2.51
130	1.83	2.07	0.14	4.04	2.27
140	1.73	2.02	0.15	3.90	2.34
150	1.64	1.97	0.15	3.76	2.19
160	1.56	1.92	0.16	3.64	2.14
170	1.49	1.87	0.16	3.52	1.97
180	1.42	1.82	0.16	3.40	1.92
190	1.36	1.78	0.16	3.30	1.84
200	1.31	1.74	0.16	3.21	1.82
225	1.19	1.64	0.16	2.99	1.70
250	1.09	1.56	0.16	2.81	1.68
275	1.00	1.48	0.16	2.64	1.53
300	0.94	1.42	0.16	2.52	1.46
325	0.87	1.35	0.15	2.37	1.35
370	0.78	1.26	0.15	2.19	1.30
400	0.73	1.20	0.14	2.07	1.12
450	0.66	1.11	0.14	1.91	1.05
500	0.60	1.04	0.13	1.77	1.06
550	0.55	0.98	0.12	1.65	0.91
600	0.51	0.92	0.12	1.55	0.87
650	0.47	0.87	0.11	1.45	0.82
710	0.44	0.82	0.11	1.37	0.77
760	0.41	0.78	0.10	1.29	0.75
800	0.40	0.75	0.10	1.25	0.70
830	0.38	0.73	0.10	1.21	0.70
860	0.37	0.71	0.10	1.18	0.68
900	0.36	0.69	0.09	1.14	0.65
1000	0.32	0.64	0.09	1.05	0.63
1100	0.30	0.59	0.08	0.97	0.58
1200	0.27	0.55	0.08	0.9	0.60
1250	0.26	0.53	0.08	0.87	0.53

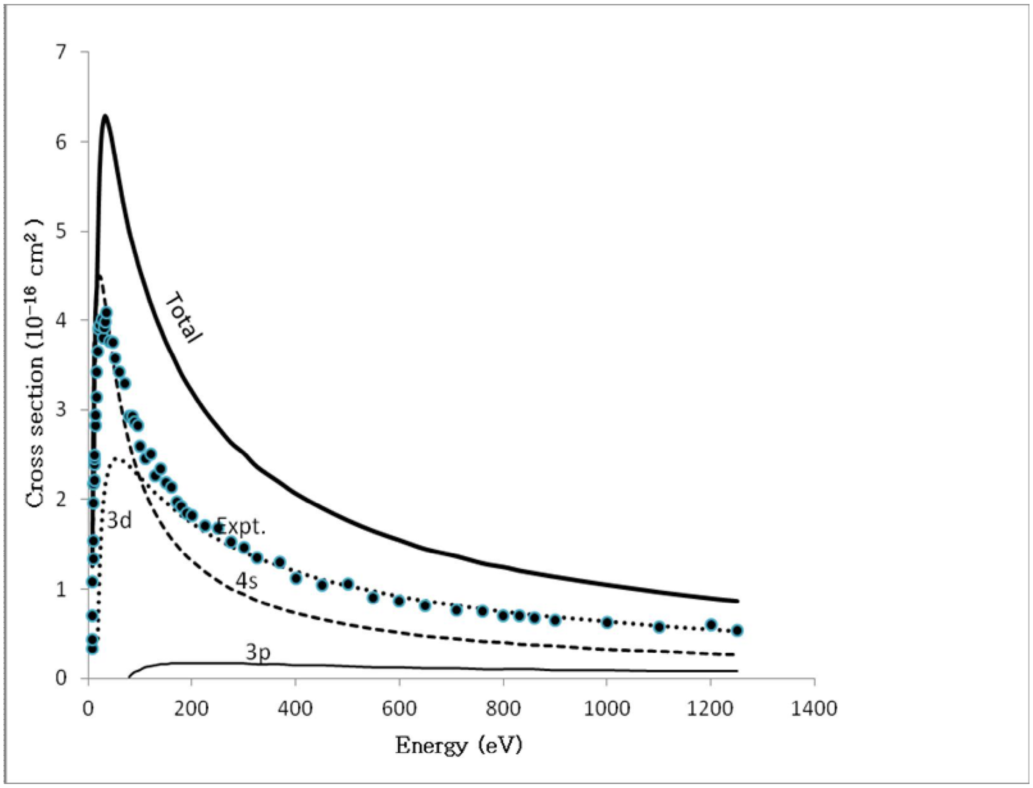

Figure 1: Electron impact single ionization cross section of Fe:
The 4s, 3d and 3p stand for the ionization cross sections for the respective shells and **Total** stands for total theoretical single ionization cross section and experimental data as **Expt.[2]**

Electron impact double ionization of Fe

The Fe has an electronic structure with 6 electrons in 3d and 2 electrons in 4s outside the close core. The experimental double ionization cross sections after attaining maxima at energies 80 eV, 90 eV, 110 eV, 140 eV,170 eV, 200eV, 250 eV and 325 eV respectively it is quite surprising that the experimental data of electron impact double ionization measured by Shah et al. [2] has eight peaks between the energy range 80 eV to 325 eV.It indicates that there are large fluctuations in the results of experimental measurements. When experimental results attains maxima at 80 eV there is slow decrease in energy in the region 80 – 325 eV indicating significant contribution from direct process. Obviously direct double ionization of Fe is considered to the result from ejection of loosely bound 3d and 4s electrons. In addition we have considered ionization of 3s electrons to lead to an exited state which results double ionization through auto ionization. The theoretical results of double ionization cross sections along with experimental data in the energy region from threshold to 1200 eV have been presented in the Table2 and Figure 2. First we would like to discuss the degree of agreement of the calculated direct double ionization results with the experimental data. Near to the threshold at impact energies 18.3 to 35.3 eV the ratios of calculated cross sections to the experimental data are always within the factor 2. After the inclusion of (4s,3d) contribution beyond the energy 40 eV the calculated cross sections and the experimental data are diverging very rapidly and the theoretical results are 4.13, 4.65, 3.97 and 3.64 times at impact energies 45 eV, 55 eV, 65 eV, and 75 eV respectively. The diversity of the calculated results along with the experimental findings might be due to the inclusion of contribution of (4s, 3d) shells in the calculations. Beyond the energy 75 – 225 eV impact energies our calculated results are more than three times and 4 times compared to the experimental data. After the impact energy 275 eV the result comes closer to each other. At this energy 760 eV the magnitude of the theoretical and experimental results are 1.04 x 10^{-17} $cm^2$and 1.02 x 10^{-17} $cm^2$and between the energy 600 eV to 800 eV the experimental results are almost flat. Beyond the energy760 eV the experimental results overestimates the calculated cross section at all impact energies up to 1200 eV. The variations of the theoretical as well as experimental results are almost similar in nature except for a few energy regions.

Table 2: Electron impact double ionization cross section of Fe in the unit of $10^{-17}cm^2$

E (eV)	Contribution of (4s,4s)	Contribution of (4s,3d)	Contribution of (4s,3p)	Contribution of 3s	Total	Expt[2]
28.3	0.27				0.27	0.28
33.3	0.48				0.48	0.61
35.3	0.54	0.49			1.03	0.74
40	0.62	2.45			3.07	0.97
45	0.66	4.26			4.92	1.19
50	0.67	5.47			6.14	1.39
55	0.65	6.2			6.85	1.46
60	0.63	6.6			7.23	1.58
65	0.61	6.79			7.40	1.86
70	0.58	6.83			7.41	1.88
75	0.55	6.78			7.33	2.01
80	0.52	6.66			7.18	2.18
85	0.49	6.51			7.00	2.11
90	0.47	6.33			6.80	2.15
100	0.42	5.95	0.34		6.71	2.33
110	0.38	5.56	0.78		6.72	2.38
120	0.34	5.19	1.07	0.02	6.62	2.35
130	0.31	4.84	1.26	0.05	6.46	2.38
140	0.28	4.52	1.38	0.07	6.25	2.36
150	0.26	4.23	1.44	0.01	5.94	2.20
170	0.22	3.72	1.48	0.11	5.53	2.07
180	0.2	3.5	1.46	0.12	5.28	2.24
190	0.19	3.39	1.44	0.13	5.15	1.77
200	0.18	3.11	1.42	0.14	4.85	2.01
225	0.15	2.72	1.32	0.15	4.34	1.91
250	0.13	2.39	1.23	0.16	3.91	1.72
275	0.11	2.13	1.13	0.17	3.54	1.78
300	0.1	1.9	1.05	0.17	3.22	1.77
325	0.09	1.71	0.97	0.17	2.88	1.78
375	0.08	1.42	0.83	0.17	2.50	1.61
400	0.07	1.31	0.78	0.17	2.33	1.54

450	0.06	1.11	0.68	0.16	2.01	1.32
500	0.05	0.96	0.59	0.16	1.76	1.31
550	0.04	0.84	0.52	0.16	1.56	1.22
600	0.04	0.74	0.47	0.15	1.46	1.08
650	0.04	0.66	0.42	0.15	1.27	1.08
710	0.03	0.58	0.38	0.14	1.13	1.07
760	0.03	0.53	0.34	0.14	1.04	1.02
800	0.03	0.49	0.32	0.13	0.97	0.98
850	0.02	0.45	0.29	0.13	0.89	0.92
900	0.02	0.41	0.27	0.13	0.83	0.89
1000	0.02	0.35	0.23	0.12	0.72	0.8
1100	0.02	0.31	0.2	0.11	0.64	0.77
1200	0.02	0.29	0.17	0.1	0.58	0.75

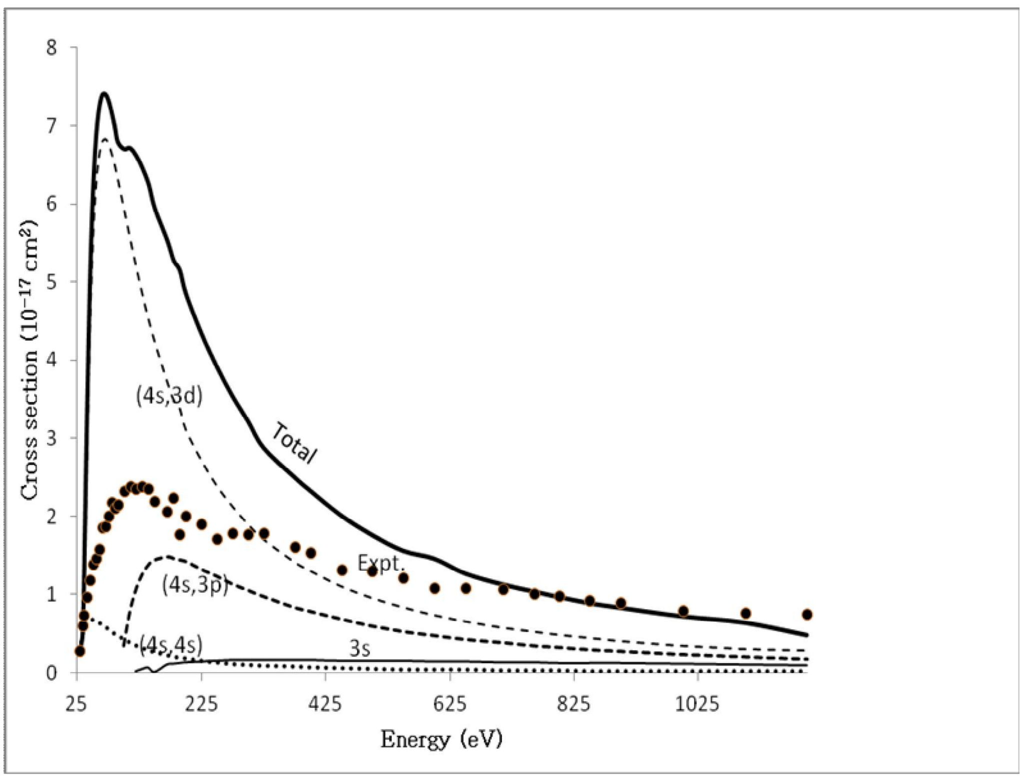

Figure 2: Electron impact double ionization cross section of Fe:

The (4s,4s), (4s,3d) and (4s, 3p) stand for partial contribution to the electron impact direct double ionization of Fe and 3s is included as an additional contribution and **Total** stands for the total theoretical double ionization cross section of Fe and experimental data as **Expt.[2]**

Our calculated results exhibits only one pick at the impact energy 70 eV having magnitude 7.41 x 10^{-17} cm^2 and indirect contributions from the inclusion of shells play an important role at high energy regions. The position of the calculated peak is found to be shifted in lower energy side as compared to the experimental counterpart. Magnitudes of cross sections above 140 eV shows a satisfactory agreement with experimental data but there is same discrepancies in the high energy regions where the calculated cross sections are found to be smaller and smaller as compared to the experiment with increase of energy. This discrepancy reflects the possibility of some other physical process contributing double ionization. Structure in the experimental double ionization cross section- curves between energy 80 eV to 325 eV attributes to indirect ionization process arising from inner shells. However, there is no indication of structure in the double ionization curve. But decrease in experimental cross section is rather slow in this energy region. This is not in accordance with the usual trend of direct double ionization cross section which shows a faster decrease in high energy region after attaining the maximum energy. Such a trend exhibits in the calculated cross section when attaining the maximum value 7.41×10^{-17} cm^2 at impact energy 70 eV, but beyond this energy the contribution of direct double ionization falls rapidly and it is observed that at the highest energy of 1200 eV the magnitude of the cross section is 0.58×10^{-17} cm^2. This means the ratio of these two calculated results of cross sections at energies 70 eV and 1200 eV is more than 12 times. Such a feature is not observed in the double ionization cross section in the measured data. Further it is suggestion for the experimentalist that there needs further experiment to perform.

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