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Application of Binary Encounter Approximation and Electron Impact on Ionization of Atoms

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ABSTRACT

Semi-classical binary encounter approximation has been used for theoretical calculations of electron impact single ionization cross sections of Xe and Kr and double ionization of Fe atom at ground state. An accurate expression of cross section for energy transfer ΔE ($\sigma_{\Delta E}$) as given by Vriens and quantum mechanical Hartree-Fock velocity distributions for target electron have been used in the calculation. In the case of single ionization of Xe, 71% theoretical results lie within valid range of ratio factor less than 2 and 55% have values less than 1.5. In the case of Kr, 97% of results have ratio factor less than 2 and 34% of results have ratio factor less than 1.1. Gryzinski and Kune model of charged particle impact double ionization of atoms found suitable for describing double ionization of atoms and ions. In the case of double ionization of Fe by electron impact 47% of results have ratio factor less than 2 and 18% of results have ratio factor 1.2. At impact 760 eV,800 eV and 850 eV having ratio factor 1.01, 0.989 and 0.967. Major contribution in double ionization is 60.16% from (4s,3d) and 31% is from (4s,3p).

Keywords: Binary encounter approximation, single and double ionization, ionization cross section, HF momentum distribution function, Vrien's accurate expression of energy transfer

1. INTRODUCTION

The collision of charged particle with a gas atom may result in a number of effects. The discovery of X rays by Rontgen in 1895 marked the beginning of quantitative studies of ionized gases. In 1952, a book by Massey and Burhop [1] appeared on as electron and ionic impact phenomenonwhich provided important knowledge for the growth of physics, both in development of experimental apparatus and theoretical development. Because of the advanced experimental set up there observed serious shortcoming between the experimental data and previously accepted theoretical methods that explains scattering and ionization of particular atoms and molecules. The ultimate goal of any theoretical scattering calculation is

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to produce accurate scattering amplitudes which can be used to predict physical observables for the system. In the past, binary encounter approximation (BEA) has been used successfully to calculate charged particle impact single and multiple ionization cross sections for atoms and ions. The process of electron-impact double ionization of atoms (e^- + X =3 e^- + X^{2+}) is very difficult to describe theoretically since the interaction of the incident electron with the atomic electrons has a collective character. Gryzinski and Kune [2] presented a general model of electron-impact double ionization of atoms that have atomic numbers $Z \geq 20$. He derived general expression for electron impact double ionization cross sections of atoms using the formalism of the binary encounter approach.

Studies of scattering theory and theory of ionization are used in different fields of science like astronomy, atmospheric physics, nuclear physics, particle physics, plasma physics, medical science, ion beam technology, and many more. The theoretical knowledge of scattering and continuous data of ionization cross sections of respective processes of atomic collision by light and heavy charged projectile($H^{=}$,He^{2+}) have great importance in different fields of science like (i) in the study of planetary atmospheres where atoms and molecules are constantly irradiated by electrons, cosmic rays and fast ions and electrons affecting their molecular inventory [3,4] and in the study of upper atmosphere where a large number of elements, both in natural and ionic forms exist and phenomenon like electron capture, transfer ionization processes are relevant for research. (ii) cancer therapy [5], where the fragmentation of water molecules present in the human body by some ionizing agent can lead to several reactive radicals that can produce local biological damages near the tumor and help in the treatment. Monte Carlo simulations track structure is usually used in micro and nano-dosimetry to find radiation transport index in medical science. Better the results of cross sections used as simulation codes better the treatment in medical science. Projectile particles of ions like protons (H^+) and helium (He^{2+}) deposit a large amount of their energy in a volume of a few micrometers or even nanometers and cause extensive damage to the microscopic structure of the biological matter and results cell death in the DNA [7, 6] and (iii) plasma physics, during the last years the use of physical plasma has been grown rapidly for medical purpose and remained as an innovative and emerging field. For the same it has an application in the human or animal body to realize therapeutic effects [7,8]

2. GENERAL THEORETICAL APPROACH

Various theoretical approaches like pure classical binary encounter approximation, Semi classical binary encounter approximation, and semi-empirical and pure quantum mechanical approximation are being used since last few decades. Different approximations have been their own limitation regarding impact energy and nature of atoms/ions. So not a particular theoretical formalism is applicable for different channels in collision problems for all range of energy. Several quintal approximations have been successfully developed for calculation of electron impact single and double ionization cross sections of light atoms/ions. In recent past quantal calculations of single and double ionization cross section of multi-electron systems have become available in the literature by Pindzola et al. [9-11].

Theoretical calculations based upon quintal treatment are very sophisticated and it fails to calculate the cross sections in case of heavy atoms. As degree of ionization increases, the problem becomes complicated in solving the wave functions quantum mechanically and the BEA can be considered to provide a suitable theoretical description of single and multiple ionization process. In the past, Semi-classical BEA has been used successfully in the calculations ofsingle and multiple ionization cross sections for several atoms and ions. The model has been used by Roy et al. [12], Kumar et al. [13], Gupta et al. [14,15,16] and Shantosh et al. [17], Singh et al. [18], Minakshi et al. [19]. Because of the improvement of classical model and inclusion of quantum theory, the Semicl-classical BEA has been used widely these days.

According to Rudge [20] three basic assumptionsare used in treating electron impact ionization in classical methods.

a. In the case of electron impact ionization, the initial state of the bound electron of target atom is either supposed to be at rest or to have a fixed velocity or to have some prescribed velocity distribution.
b. Collision processes aredescribed as though it were a two body like Binary Encounter Approximation(BEA). Theoretical results of ionization cross section are supposed to be valid if ratio factor is less or equal to 2. The binary encounter model is based on the following two assumptions [21]:

1. Electrons of the target atom are regarded as completely independent of each other in course of collision. The momentum transferred by the incident particle to one of the target electron would be larger than the momentum with orbitalelectrons and energy transferred would be much larger than binding energy of the bound electron.

2. Prior to the collision, target electrons are regarded as free particles having a velocity distribution.

Electron impact single and double ionization of light and heavy atoms or ions have been discussed below.

Electron impact single ionization cross section

Thomson first used the binary encounter theory for calculating cross section for ionization of atom by electrons. Thomson considered a situation of collision where the energy transfer in Coulomb collision between a particle of mass m_1 and charge Z_1e with initial kinetic energy E_1 and a particle of mass m_2 and Z_2e with initial kinetic energy $E_2 = 0$ (rest). The

Thomson's energy transfer (ε) ionization cross section for electron –electron collision is [21]

$$\frac{dQ(\varepsilon)}{d\varepsilon} = \pi \frac{e^4 N}{E_1}\left[\frac{1}{U} - \frac{1}{E_1}\right]$$

(1)

The atom gets ionized if, $U \le \varepsilon \le E_1$; where U is ionization potential energy.

Thomas and William (1927) modified the formulation for more general case where $E_2 \neq 0$ (considered symmetrical distribution of velocity of target electrons), $m_1 \rangle\rangle m_2$ and $Z_1 \neq Z_2$ which is relevant to proton and alpha particle –atom collision. Energy transfer ionization cross section for this case has been given as [22]

$$\frac{dQ(\varepsilon)}{d\varepsilon} = \pi \frac{e^4 Z_1^2 Z_2^2 m_1}{m_2 E_1}\left[\frac{1}{\varepsilon^2} + \frac{4E_2}{3\varepsilon^3{}_1}\right]$$

(2)

These classical theories remained dormant for three decades after pioneering work of Gryziniski [23] in the literature. New progress was made by Gryziniski and obtained classical relations for Coulomb collision of two moving charged particles and applied them for theoretical studies of a varity of charged particle- atom collision processes. Variens [24] gave a set of quantum mechanical formulas for scattering of one electron by another interms of momentum transfer as a variable. He incorporated symmetrical properties in the formulation that includes effects exchange and interference and obtained differential cross section for momentum and energy transfer.

The theory has been successfully employed, improved and extended by many workers leading to modified semi-classical binary encounter approximation. Expression for electron impact single ionization [24],

$$Q^i = \frac{\pi e^2}{E_1 + E_2 + U}\left\{\left(\frac{1}{U} - \frac{1}{E_1}\right) + \frac{2E_2}{3}\left(\frac{1}{U^2} - \frac{1}{E_1^2}\right) - \frac{\phi^i \ln\left(\frac{E_1}{U}\right)}{E_1 + U}\right\}$$

(3)

where

$$\phi^i = \mathrm{Cos}\left[\left(\frac{R}{E_1+U}\right)^{\frac{1}{2}} \ln\left(\frac{E_1}{U}\right)\right]$$

(4)

E_1 and E_2 are kinetic energies of incident and orbital electron of the target respectively; R and U are Rydberg constant and the ionization potential of the shell under consideration where $U = v_0^2$.

The above expression has been rewritten in terms of two well-known dimensionless quantities s and t defined as [25] $s^2 = \frac{v_1^2}{v_0^2}$ and $t^2 = \frac{v_2^2}{v_0^2}$; v_1, v_2 and v_0 are velocities of the incident, bound electron and mean orbital velocity respectively. All other energies have also been expressed in Rydberg. $E_1 = ½ m_1 v_1^2 = ½ m_1 s^2u$, for electron $m_1=1$ and $E_2 = ½ m_e v_2^2 = ½ t^2u = t^2u$. In terms of these dimensionless quantities, equation (2) takes the form

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

(5)

where

$$\phi^i = \left[\left(\frac{1}{s^2U+U}\right)^{\frac{1}{2}} \ln s^2\right]$$ (6)

Impact energy gets distributed among orbital electrons and to include such effects an analytical expression of Hartree-Fock velocity distribution function ($f(t)$) is multiplied with $Q_i(s,t)$ and integrated to give total single ionization cross section of a particular atom

$$Q_i(s) = n_e \int_0^{\alpha} Q_i(s,t)\, f(t)\, U^{\frac{1}{2}} dt \quad (7)$$

where n_e is the number of equivalent electrons in the shell.

Here $f(t)$ is constructed with the following quantum mechanical expressions[7].

$$f(t) = 4\pi t^2 U \rho_{nl}(U^{1/2}t) \quad (8)$$

in which

$$\rho_{nl} = \frac{1}{2l+1}\sum_{-1}^{+1} |\psi_{nlm}(x)|^2 \quad (9)$$

where

$\psi_{nlm}(r) = \frac{1}{(2\pi)^{\frac{1}{2}}} \int \phi_{nlm}(r) e^{ik.r} dr$ is the Fourier transform of the one electron orbital.

The complete wave function or Slater orbitals is given by;

$$\phi_{nlm}(r) = N_{nl} R_{nl}(r) Y_{lm}(\Omega) \quad (10)$$

where N_{nl}, $R_{nl}(r)$ and $Y_{lm}(\Omega)$ are the sormalization constant, Hartree-Fock radial function and spherical harmonic respectively and can be given as

$$N_{nl} = [(2n)!]^{-\frac{1}{2}} (2\xi)^{n+\frac{1}{2}} \quad (11)$$

and

$$R_{nl} = r^{n-1} e^{-\xi r} \quad (12)$$

Here zeta (ξ) is orbital exponent of basis function. The spherical harmonic $Y_{lm}(\Omega)$ have different forms depending upon the value of orbital and magnetic quantum numbers l and m respectively.Computational calculation of equation (7) finally gives results of SICS for a particular orbital under different selective constants of the respective subshell. The expression of $Q_i(s,t)$ and $f(t)$ are taken from equation (5) and (4) respectively. The momentum distribution function $f(t)$ is constructed from equations (8-12) for particular orbital electron of the target atom as discussed above.

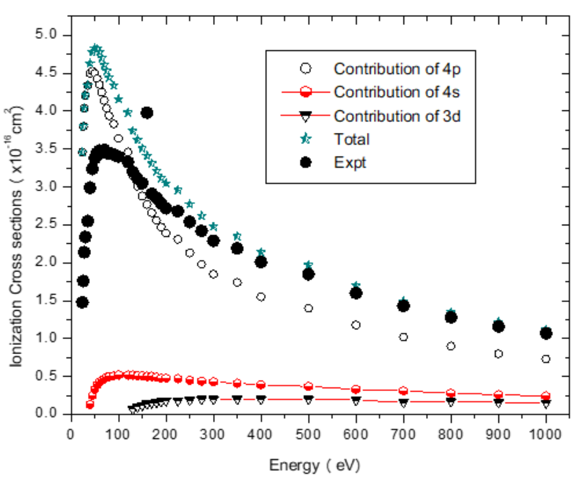

Fig. 1a: Electron impact SICS of Kr.

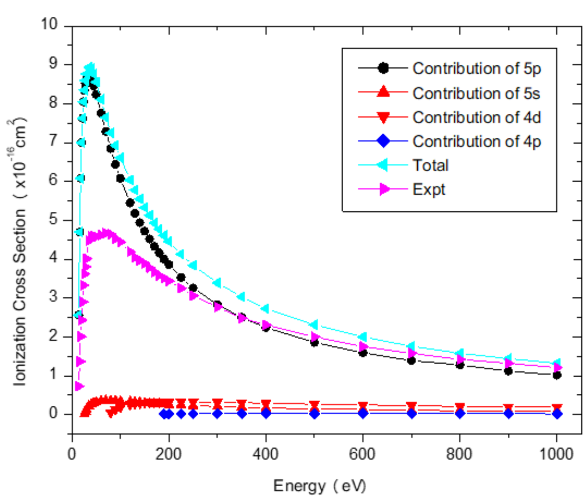

Fig. 1b: Electron impact SICS of Xe.

Fig.2: Variation of total electron impact SICS of Kr and Xe atoms [26,27]

For shell radii and binding energies of electrons, quantum mechanical value of radial distance of maximum probability given by Desclanx [28] and quantum mechanical value of orbital energies given by Clementi and Roetti [29] have respectively been used in the calculations.The mathematical formulation for single ionization cross section of atoms under semi-classical BEA formulation was able to explain very well the experimentally observed data of single ionization cross section of Kr and Xe [26, 27] as shown in Fig. 1a and Fig. 1b. According to Rudge theoretical results are considered valid if ratio factor is less or equal to two. Here ratio factor for impact energy 200 eV to 1000eV lies in between 1.1 to 1.05 (almost equal to experimental result). This shows that calculated results for proposed model is in excellent agreement with the experimental data. In the same way, the calculated values of single ionization cross section of Xe atom are also in good agreement with experimental data except in low energy range as shown in Fig. 1. In the case of single ionization Xe and Kr, 71% theoretical results of SICS of Xe have ratio factor less than 2 and 55% have values less than 1.5. In the case of Kr 97% of calculated results have ratio factor less than 2 and 34% of results have ratio factor less than 1.1. In energy range 700 eV to 1000 eV ratio factor is nearly 1.05. The ultimate and utmost goal and success of theoretical formalism for the ionization phenomena lies in its ability to explain the experimental observations. Hence, the model discussed above explains well the phenomenon of single and multiple ionization of atoms/ions and is widely used now adays.

Model Mechanism of Double Ionization

According to the idea suggested by Gryzinski and Kune [2], the double ionization of an atomic target by projectile may proceed via two alternative processes.

(i) The two electrons may be ejected from the atom by two successive encounters of the incident charged particle with the target electrons.

(ii) Alternatively, the incident particle may knock out only one electron and the second electron is removed by the second electron. The total double ionization cross section for the target is given by the sum of contributions of double ionization from the two alternative processes (Figure 2).

The electron-impact double ionization of atoms is a result of two consecutive electronic encounters. The electron-impact double ionization of an atom proceeds by two ‘direct’ and ‘indirect’, mechanisms. The ‘direct’ ionization consists of two consecutive scatterings (Figure 2a). First the incident electron e is scattered from one of the atomic ‘active’ electrons e_1 in the ‘collision region I and it results in ejection of electron e_1 from the atom.

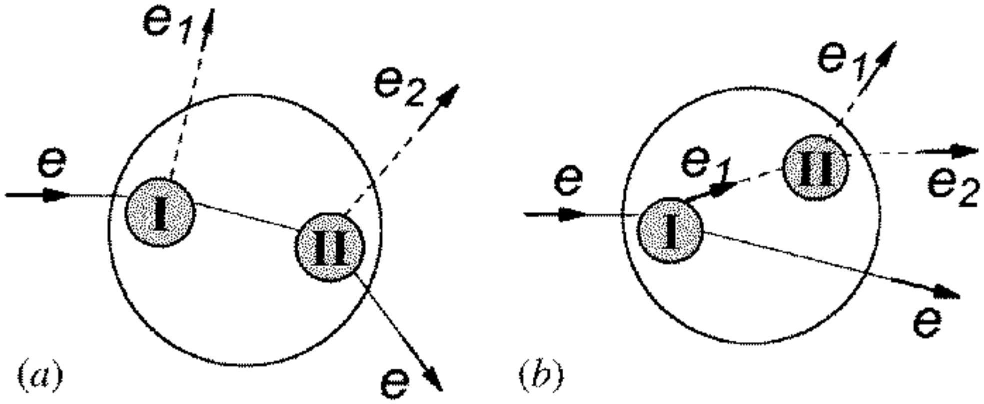

Fig. 2: Gryzinski-Kun model of double ionization: (a) The 'direct' mechanism and (b) the 'indirect' mechanism of electron impact double ionization of atoms.

The ejected electron leaves the collision region I and the scattered electron enters the 'collision region II and collides with the electron $e_2$and $e_2$comes outfrom atom. The 'indirect' double ionization also consists of two consecutivescatterings (Figure 2b). In the first scattering, the incident electron e is scattered from the active- electron e_1 in the collision region I, and moves towards the collision region II while the electron e leaves the atom. In the second scattering (taking place in the collision region II), the electron e_1 collides with the active-electron e_2 and e_2 leave the atom. The approach developed in this work applies to atoms with outer shells where the motion of the 'active' electrons is confined to s and d shells. Hartree-Fock and hydrogenic velocity distributions were used while considering the first and second target electrons respectively.

The expressions for cross sections corresponding to the above two processes of the double binary encounter model leading to direct double ionization (when Auger emission is not considered) is given by

$$Q_D^{II} = Q_{sc}^{II} + Q_{ej}^{II} \#(13)$$

where Q_{sc}^{II} and Q_{ej}^{II} are double ionizations cross sections under two alternative processes. In the first process, the two target electrons are ejected from two successive encounters by the incident electron denoted by Q_{sc}^{II}. Alternatively, the incident electron may knock out only one target electron and the second electron is removed by the first ejected electron denoted by Q_{ej}^{II} . The expressions for Q_{sc}^{II} and Q_{ej}^{II} have been integrated numerically over energy transfer and Hartree-Fock momentum distribution for ejection of the two electrons.

Electron impact double ionization cross section

The expressions for Q_{sc}^{II} and Q_{ej}^{II} given by Gryzinski [23c] and modified by Roy and Rai [30] are

$$Q_{sc}^{ii} = \frac{n_e(n_e-1)}{4\pi\bar{r}^2}\int_{U_i}^{E_R-U}\sigma_{\Delta E}\left[\int_{U_{ii}}^{E_R-\Delta E}\sigma_{\Delta E}\,d(\Delta E)\right]d(\Delta E)\#(14)$$

and

$$Q_{ej}^{ii} = \frac{n_e(n_e-1)}{4\pi\bar{r}^2}\int_{U_i+U_{ii}}^{E_R}\sigma_{\Delta E}\left[\int_{U_{ii}}^{E_R-\Delta E}\sigma_{\Delta E}\,d(\Delta E)\right]d(\Delta E)\#(15)$$

The above two expressions have been integrated numerically over energy transfers and the Hartree-Fock velocity distributions for the ejection of the two electrons under double BEM leading to direct double ionization are given by

$$Q_{sc}^{ii} = \frac{n_e(n_e-1)}{4\pi\bar{r}^2}\int_{t=0}^{\infty}\int_{U_i}^{E_R-U_{ii}}\sigma_{\Delta E}\left[\int_{t=0}^{\infty}\int_{U_i}^{E_R-\Delta E}\sigma_{\Delta E'}\,f(t)\,U_{ii}^{\frac{1}{2}}\,d(\Delta E')\,dt\right]\#$$
$$\times f(t)\,U_i^{\frac{1}{2}}\,d(\Delta E)\,dt \times 8.797\times10^{-17}(\pi a_0^2)\#(16)$$

and

$$Q_{ej}^{ii} = \frac{n_e(n_e-1)}{4\pi\bar{r}^2}\int_{t=0}^{\infty}\int_{U_i+U_{ii}}^{E_R}\sigma_{\Delta E}\left[\int_{t=0}^{\infty}\int_{U_{ii}}^{\Delta E-U_i}\sigma_{\Delta E'}\,f(t)\,U_{ii}^{\frac{1}{2}}\,d(\Delta E')\,dt\right]\#$$
$$\times f(t)\,U_i^{\frac{1}{2}}\,d(\Delta E)\,dt \times 8.797\times10^{-17}(\pi a_0^2)\#(17)$$

Here n_e is the numbers of electrons in the shell under consideration, ΔE and $\Delta E'$ stands for energy transfers during the first and second collision respectively, $\bar{r}$ denotes the mean distance between the electrons in the shell given by $\bar{r} = R/n_e^{1/3}$ where R being radius of the shell of the target. Also U_i and U_{ii} are the ionization potentials corresponding to the ejection of the two electrons of the target.Using dimensionless variables introduced by Catlow and McDowell, the accurate expression of cross section $\sigma_{\Delta E}$ (Varien, 1966) can be given as

$$\sigma_{\Delta E} = \frac{2}{(s^2+t^2+1)u}\left[\left(\frac{1}{\Delta E^2}+\frac{4t^2u}{3\Delta E^3}\right)+\left(\frac{1}{(s^2u+u-\Delta E)^2}+\frac{4t^2u}{3(s^2u+u-\Delta E)^3}\right)\right.\#$$
$$\left.-\frac{\phi}{\Delta E(s^2u+u-\Delta E)}\right]\pi a_0^2 \qquad \#(18)$$

where

$$\phi = \cos\left\{\left(\frac{1}{s^2u+u}\right)^{\frac{1}{2}}\ln s^2\right\}\#(19)$$

Due to indistinguishability of electron in the symmetrical model of Vriens the cross sections corresponding to two processes are exactly equal at all incident energies and hence in order to obtain the direct double ionization cross section, either of the cross sections should be multiplied by two. In the above equation u and s^2 have been replaced by U_i and E_q/U_i 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 symbol E_q is the energy of the projectile. The function $f(t)$ appearing in the above equations is the momentum distribution function. In case of double ionization $f(t)$ has been constructed replacing ionization potential energy u by U_i and U_{ii} for ejection of first and the second electron respectively.

For example, in the case of Fe, the total cross section for electron impact direct double ionization is given by

$$Q_D^{ii} = Q_D^{ii}(4s,4s) + Q_D^{ii}(4s,3d) + Q_D^{ii}(4s,3p) \# (20)$$

where $Q_D^{ii}(4s,3d)$ stands for the double ionization cross section corresponding to the one electron ejected from the 3d shell and other from 4s shell and $Q_D^{ii}(4s,4s)$ stands for ejection of the two electrons from same shell. Direct double ionization of Fe is considered due to ejection of loosely bound electrons from 3d and 4s sub shells. In addition, we have considered ionization of 3s electrons to lead to an exited state which results double ionization through auto ionization. I would like to discuss the degree of agreement of the calculated direct double ionization results with the experimental data. Major contribution is from (4s, 3d) and very insignificant contribution is from (4s, 4s) and (4s, 3p) as shown in Fig.3. After the inclusion of (4s, 3d) contribution beyond the energy 40 eV the calculated cross sections and the experimental data are diverging rapidly. Beyond impact energy 275 eV the results come closer to each other. At energy 760 eV the magnitude of the theoretical and experimentalare $1.04 \times 10^{-17} cm^2$ and $1.02 \times 10^{-17} cm^2$ (nearly same) and between energy 600 eV to 800 eV the experimental results are almost flat.

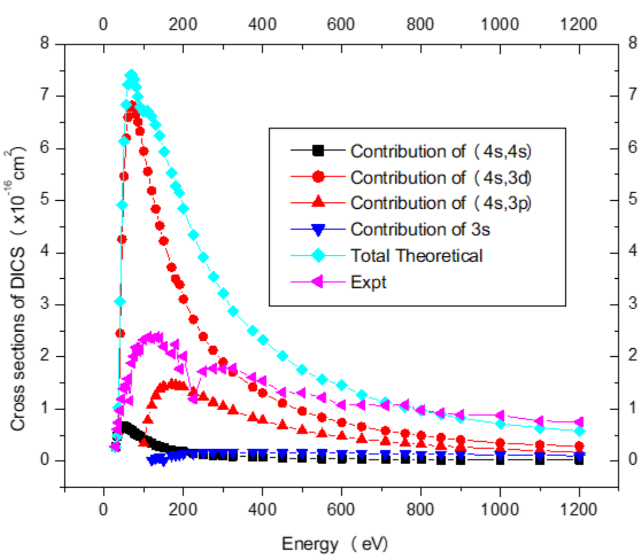

Fig. 3: Electron impact double ionization cross section of Fe atom along with its experimental values of single ionization cross section (Shah et al., 1993) [31] against respective impact energy

The variations of the theoretical as well as experimental results are almost similar in nature except at some lower energy regions. Indirect contributions from the inclusion of 3d shells play an important role at high impact energy regions. About 58% of calculated results have ratio factor less than two and 29.4% have less than 1.5 in between 300 keV/amu to 1440 keV/amu. Magnitudes of cross sections above 250 eV shows a satisfactory agreement with experimental data but there are 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 impact energy.

This discrepancy reflects the possibility of some other physical processes contributing to 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. 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.

4. CONCLUSION

Exact solutions of atomic collision processes are not possible. The validity of any theoretical approach lies in its ability to unveil the mystery of any physical phenomena experimentally observed and consequently depends upon the degree of closeness of theoretical results to the experimental results. In the case of single ionization of Xe, Kr and double ionization of Fe by electron have been discussed above. Maximum number of theoretical results are close to the corresponding experimental values. In the case of single ionization of Xe, 71% theoretical results have ratio factor less than 2 (valid reliable range) and in the case of Kr, 97% of results have ratio factor less than 2 and in the energy range 700 eV to 1000 eV ratio factor are nearly 1.05. Gryzinski and Kune (1999) model of charged particle impact double ionization of atoms found suitable for describing double ionization of atoms and ions. In the case of double ionization of Fe by electron 47% of results have ratio factor less than 2 and 18% of results have ratio factor 1.2. At impact 760 eV, 800 eV and 850 eV having ratio factor 1.01, 0.989 and 0.967. Major contribution in double ionization is 60.16% from (4s, 3d) and 31% is from (4s, 3p). Above discussion of theoretical results using binary encounter approximation are in favor of application of the semi-classical model. Also this model is used by a number of workers to explain experimental results for proton and alpha particle single double ionization of different light and heavy atoms atoms and ions.

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