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He^{2+} impact single ionization cross sections of Cu atom

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ABSTRACT

Semi-classical binary encounter approximation has been used for theoretical calculations of single ionization cross sections of Cu atom at ground state by alpha particle impact in energy range varying from threshold 35 keV/amu to 425 keV/amu. 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 electrons have been used in the calculation. Major contribution to the total single ionization cross sections of Cu are from 4s and 3d subshells. The ionization cross sections decrease with the increase of impact energy same as experimental data reported. The theoretical and experimental results of single ionization cross sections have same trends against the increase of impact energy. The ratio factor falls within 2, varying from 1.26 to 1.86, for given energy range. Theoretical results are under valid range. About 50% of total theoretical results of single ionization cross sections have ratio factor (R) ≤ 1.5. Major contribution to total ionization cross section is from 3d and 4s subshells-electrons whose contributions varies from 62 to 71% and 41.7% to 23.8% respectively. The higher value of linear correlation coefficient (=0.9647) and lower value of standard deviation (=0.822) shows that results calculated are close to the experimental data in the intermediate and high energy range.

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1. Introduction

Single and multiple ionizations of atoms and molecules by ionizing particles like electrons and ions is one of the fundamental processes in atomic physics. Collision between heavy charged particles (H^+ or He^{2+}) with target atoms may results pure ionization, excitation, excitation-auto-ionization, electron capture, charge transfer and transfer ionization. Molecular dissociation is a widespread phenomenon that can produce ions, atoms and molecular radicals which are far more reactive than the original molecules. Molecular dissociation comprises a molecular medium and some kind of ionizing radiation. Continuous data of ionization cross sections of respective processes have great importance in different fields of science. Some important and quite distinct examples of this combination are (i) planetary atmospheres that are constantly irradiated by electrons, cosmic rays and fast ions affecting their molecular inventory [1-4]. A large number of elements both in natural and ionic forms exist in the upper atmosphere and the capture processes are relevant to upper atmosphere research. From astrophysical point of view, the charge exchange in alpha particle- atom collision is important because the emission spectrum of solar chromosphere contains spectral line $\lambda = 4686\ A^0$ whose origin has been attributed to the presence of ionized helium formed due to the process of electron capture by fast alpha particles produced in nuclear reaction [5] (ii) cancer therapy [6,7] 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 is 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 biological matter and results cell death in the DNA and (iii) plasma physics, where the environment of ions reacting with each other have many applications, such as plasma etching of microchips [8]. Indirectly the plasma-supplemented techniques are used to treat surfaces, materials and some devices to realize specific qualities. Physical plasma has an application in the human or animal body to realize therapeutic effects [9,10]. Therefore, a set of continuous and precise data of single and multiple ionization cross sections of different atoms are of great importance in the study of different fields of science as mentioned above. So, the theoretical results of ionization cross sections of different atoms have their own importance in physics.

Multiple-ionization is a complex many-electron process where direct and indirect ionization contribute to the final charge state. Pindzola et al.[11] time-dependent close-coupling method in spherical polar coordinates is developed to calculate the electron-impact double ionization of the H_2 molecule. Montanan et al. [12] investigated multiple ionization of Ar by impact of alpha particle using quantum mechanical model of continuum distorted wave ekonal initial state (CDWEIS). The theoretical results investigated were quite reasonable with experimental data at high energies range. Despite these successes, difficulties still exist in the mathematical formulation for the calculation of single and multiple ionization cross sections of heavy atoms under quantal approximations.

Since the beginning of nineteenth century semi-classical theory is being used successfully along with its gradual modification. Heavy charged particles (H^+, He^{2+}) impact direct single and double ionization cross sections of different atoms have been investigated theoretically using modified binary encounter approximation by Singh et al [13], Minakshi et al. [14] Tan et al 1981, Kumari et al. [16] and Gupta et al. [17] to calculate direct single and double ionization cross sections of several light and heavy atoms/ions by the impact of heavy charged particles.

2. Methods and theoretical details

Thomson first used the binary encounter theory for calculating cross section for ionization of atom by electrons. According to Thomson consider 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). In the case of binary encounter theory, it has been assumed that during the period of interaction between projectile and an orbital electron the other atomic electrons and the nucleus play no role. The Thomson's energy transfer (ε) ionization cross section for electron –electron collision is [18]

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

For ionization $U \le \varepsilon \le E_1$; where N is the effective number of electrons in the atom and 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), $m_1 >> m_2$ 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 [18]

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

These classical theories remained dormant for three decade till the pioneering work of Gryziniski [19]. in the literature. New progress was made by Gryziniski. He 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. Gryzinski solved problem of collision using scattering angle insteed of momentum transfer as a variable. Variens [20] gave a set of quantum mechanical formula for scattering of one electron beam by another interms of momentum transfer as a variable. He incorporated symmetrical properties in the formulation that includes exchange and interference effects and obtained differential cross section for momentum and energy transfer.

We carry out theoretical calculations of alpha particle (He^{2+}) impact single ionization cross sections of Cu atom using the modified BEA. The theoretical approach used in BEA is based on independent particle model (IPM). The model is based on the hypothesis that the probability of ionizations is directly related to the energy deposited by the projectile on the target. The energy deposited is statistically distributed among all atomic electrons and one or more of which eventually auto ionize to the final state. An accurate expression of $\sigma_{\Delta E}$ (cross section for energy transfer ΔE) for proton impact given by Vriens [21] and quantum mechanical Hartree –Fock velocity distribution functions for bound electrons of the target atoms or ions have been used to calculate total single ionization cross section of iron.

Following McDowell [22], Catlow and McDowell [23] gave an expression of single ionization cross section of an atom by an electron and proton impact in terms of dimensionless variables s and t. The variables are related to kinetic energies of incoming and orbiting electrons and defined as $s^2 = v_1^2 / v_0^2$ and $t^2 = v_2^2 / v_0^2$, where v_1 and v_2 are the velocities of incident particle and target orbiting electron in atomic units respectively and v_0 is root mean square velocity of orbital electron. The ionization potential energy of bound electron u is defined as $u = v_0^2$. Atomic electrons are taken to have a momentum distribution and can be given by Fourier transformation of the Hartree - Fock density distribution that includes quantum-mechanical velocity distribution for the bound electrons. Following Catlow and McDowell, the expression of total single ionization cross section for heavy charged particle impact having energy of $m_1 s^2 u$ with an orbital electron of a particular shell having energy $t^2 u$ is given by

$$Q_i(s) = n_e Z^2 \int_0^\infty Q_i(s,t)\, f(t)\, u^{1/2}\, dt\, (\pi a_0^2) \qquad (3)$$

Where *Q(s)* is total single ionization cross section, n_e is the number of electrons in the shell under consideration, Z is the charge on the projectile (for proton and electron Z = 1 and 2 for alpha particle), $f(t)$ is Hartree-Fock momentum distribution function and a_0 is Bohr's radius. In the present calculations, $Q_i(s,t)$ is calculated using an accurate expressions of differential cross section $\sigma_{\Delta E}$ (cross section for energy transfer ΔE) under three different limits of energy transfer as given by Vriens [21]

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

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

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

Integration over differential cross section in the above three cases of energy transfer gives $Q_i(s,t)$ for the impact of unit heavy charged particle in terms of dimensionless variables as

$$\begin{aligned} Q_i(s,t) &= \frac{4}{s^2u^2}\left[1 + \frac{2t^2}{3} - \frac{1}{4(s^2-t^2)}\right]; \quad 1 \leq 4s(s-t) \\ &= \frac{2}{s^2u^2t}\left[\frac{1}{4(s+t)} + t + \frac{2}{3}\left\{2s^3 + t^3 - (1+t^2)^{3/2}\right\}\right]; \quad 4s(s-t) \leq 1 \leq 4s(s+t) \\ &= 0; \quad 1 \geq 4s(s+t) \end{aligned} \tag{5}$$

The numerical integration of $Q_i(s,t)$ carried out over Hartree-Fock momentum distribution function $f(t)$ of the bound electron that yields total ionization cross section $Q_i(s)$ [equation (3).

The momentum distribution function $f(t)$ is defined as,

$$f(t) = 4\pi t^2 u \rho_{n6}(u^{1/2}t) \tag{6}$$

where

$$\rho_{nl} = \frac{1}{2l+1}\sum_{-1}^{+1}|\psi_{nlm}(x)|^2 \tag{7}$$

and

$$\psi_{nlm}(r) = \frac{1}{(2\pi)^{\frac{1}{2}}}\int \phi_{nlm}(r)e^{ik.r}dr$$

is the Fourier transform of the one electron orbital. The complete wave function is given by

$$\phi_{nlm}(r) = N_{nl}R_{nl}(r)Y_{lm}(\Omega) \tag{8}$$

where N_{nl} and $R_{nl}(r)$ are the normalization constant and analytical Hartree-Fock radial function, respectively. The empirical relations for N_{nl} & $R_{nl}(r)$ are

$$N_{nl} = [(2n)!]^{-\frac{1}{2}}(2\xi)^{n+\frac{1}{2}} \tag{9}$$

and

$$R_{nl} = r^{n-1}e^{-\xi r} \tag{10}$$

Here 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. It is well known that velocity of orbital electrons increases with the decrease in shell number and hence electron of inner shell possess relativistic in nature. Here we have ignored the relativistic nature of orbiting electron. In the present work, ionization from valence shells and few inner shells have only been considered since rest inner orbitals have negligible contribution to the ionization cross sections. In the mathematical formulation of BEA there used non-relativistic wave functions.

3. Results and Discussion

Computational calculation of equation (3) 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 (6) respectively. The momentum distribution function $f(t)$ has been constructed from equations (7-10) for particular orbital electron of the target atom as discussed above. For shell radii and binding energies of electrons, quantum mechanical value of radial distance of maximum probability given by Desclanx [24] and quantum mechanical value of orbital energies given by Clementi and Roetti [25] have respectively been used in the calculations.

We have considered contributions only from 4s, 3d and 3p subshells as inner shells have negligible effect. Theoretical investigation of direct single ionization has been carried out in the energy range of 35 keV/amu to 425 keV/amu (Patton et al.[26]) using BEA. We compared the theoretical result of SICS with the experimental data of single ionization cross sections for corresponding impact energy. The computational calculation includes contribution of 4s, 3d and 3p orbitals. Theoretical results of SICS of these orbitals and experimental data against corresponding impact energies have been presented in Table 1 and Fig. 1.

Table1: Alpha particle impact SICS of Cu atom for different impact energies.

E (keV/amu)	Contribution of			Total SICS ($\times 10^{-16}$ cm^2)	
	4s	3d	3p	Theory	Expt. [26]
35	6.02	7.6	0.06	13.70	22.7±1.5
40	5.45	7.6	0.07	13.18	24.0±2.0
47	4.81	7.6	0.09	12.57	20.1±1.2
54	4.34	7.6	0.10	12.06	21.5 ±1.3
62	3.92	7.7	0.12	11.79	18.5 ±1.0
75	3.42	7.2	0.14	10.77	15.8 ±1.2
88	3.04	6.9	0.16	10.13	15.0 ±1.3
108	2.67	6.5	0.18	9.39	13.7 ±1.0
125	2.34	6.0	0.20	8.60	11.5 ±0.2
150	2.03	5.4	0.22	7.74	9.8 ±0.6
180	2.75	4.9	0.24	7.95	10.3 ±0.5
213	1.52	4.3	0.25	6.16	8.5 ±0.4
250	1.32	3.8	0.25	5.45	8.2 ±0.4
300	1.12	3.2	0.25	4.65	7.2 ±0.3
360	0.94	2.7	0.24	3.92	6.9 ±0.3
425	0.80	2.3	0.23	3.35	6.2 ±0.4

The theoretical and experimental results of ionization cross section have the same trend against increase of impact energy. The experimental observations overestimate the theoretical results of SICS of Cu for all given energies. The variation of the results in both cases is almost four times. The theoretical results decrease very slowly with the increase of impact energy. Except 54 and 180 keV/amu, values of experimental data calculated and experimental results of SICS of cross sections decreases with impact energies. From a number of theoretical works done by Percival (1966), Vriens (1966) and Rudge (1968), binary encounter model gives reasonable formula that estimates ionization cross sections over a significant range of energies if the ratio factor (theoretical result to the corresponding experimental value) is less or equal to 2. Here, in our case our results have ratio factors (R) falls within 2 for all given energy values. It varies from 1.26 to 1.86. This shows that all the results are within valid range. About 50% of theoretical results of SICS have ratio factor (R) ≤ 1.5 As shown in the Table 1 major contribution to the total theoretical results are from 3d shell which varies from 62 to 71% . In the same way 4s has contributions varying from 41.7% to 23.8% and 3p has very small contribution varying from 0.51% to 6.8% for entire energy range. Also, the variations of theoretical and experimental values of SICS at low to high impact energies are 4 and 3.66 respectively. We observed that the contribution of 3p is very small compared to the 4s and 3d subshells. It is found that 3p electrons have lower energy compared to 3d and 4s subshells in the electronic configuration of Cu. Only those electrons take part in pure ionization whose energies are high. Here subshells 4s and 3d have greater energies compared to 3p and nest lower subshells. According to Aufbau principle 4s is filled first if 3d has no electron. As 3d get populated with electrons, the relative energy of 4s and 3d fluctuate relative to one another and 4s ends up with higher energy state and ionization results from 3d and 4s of Cu. The nature of variation observed in theoretical results is nearly same as that of variation in experimental data and all the theoretical results have ratio factor less than 2. This shows that these theoretical results are close to the corresponding experimental data.

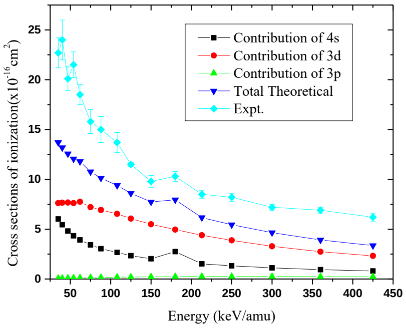

Fig.1: Alpha particle impact SICS of Cu atom in the given energy range

The Model does not include all physical insight of ionization at low energy range. The sharp fall in single ionization cross sections of 4s in threshold energy range is due to lack of suitability of our semi-classical model of binary encounter approximation.

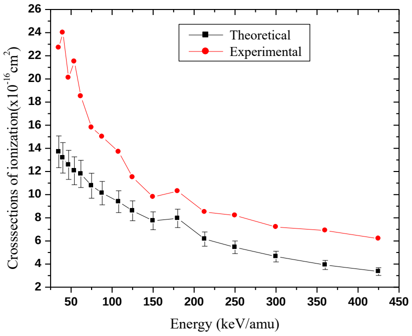

Fig. 2: Error bars associated to the theoretical results relative to the experimental data.

The variation of error associated with theoretical results in comparison with corresponding experimental values has been shown in Fig. 2. The errors associated with theoretical results have relatively high values at low energies and decreases with the increase of impact energies.

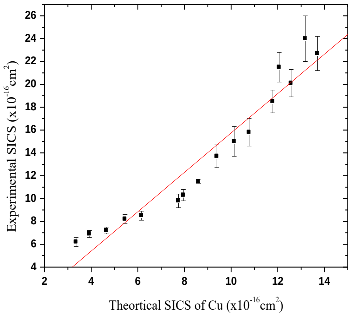

Fig.3: Linear fit for the theoretical results with the experimental data along with error-bars.

Fig.3 shows that linear correlation coefficient is 0.9647 and standard deviation (SD) is 0.822. This shows that about 96% of theoretical data are in close agreement to the line of best fit. In threshold energy range the theoretical results are more apart from corresponding experimental data and possess relatively more error compared to higher energy region. Smaller value of standard deviation shows that the theoretical results are close to the experimental values in intermediate and high energies.

Conclusion

There observed that He^{2+} impact single ionization cross sections of Cu are well explained by considering direct ionization of 4s, 3d and 3p subshells. All the theoretical results have ratio factor below two and nature of variation is nearly same as of the experiment. As discussed earlier the ratio factors falls within two for all given energy values. It varies from 1.26 to 1.86. About 50% of theoretical results of SICS have ratio factor (R) ≤ 1.5 as shown in the Table 1. The major contributions to the total theoretical results are from 3d subshell which varies from 62 to 71% and 4s has contributions of 41.7% to 23.8%. Fig.2 shows that theoretical results are close to the respective experimental values in the intermediate and high energy range and Fig.3 shows that 96% of theoretical results are in close and satisfactory agreement with the experimental data for wide range of impact energies.

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