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DOUBLE IONIZATION OF COPPER BY ELECTRON IMPACT

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Electron-impact double ionization of copper has been investigated in the modified double-binary-encounter model. For the first time, Hartree-Fock velocity distribution has been used while considering ejection of both electrons from the target in binary-encounter calculations. Contribution of Auger emission, which is considered to be dominant at high impact energies, is also included in the calculations. The present results have been compared with the recent experimental measurement of Bolorizadeh et al. and semi-empirical calculations of Belenger et al. The semi-empirical calculations are found to overestimate the cross-sections. The present results are in satisfactory agreement with the experimental observations.

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1. Introduction

Multiple ionization arising in electron-atom/ion collisions is one of the most fundamental collision processes in atomic and molecular physics. This process finds important applications in plasma kinematics problems, planetary upper atmosphere, mass spectrometry etc. In addition, it is of great interest for our understanding of the many-electron processes. Thus, these studies are very important from both experimental and theoretical points of view.

In recent past, accurate experimental measurements of multiple ionization of metallic species [1,2] have been carried out. It may be noted that many metallic species are important in fusion-energy research and astrophysical problems. More recently, Bolorizadeh et al. [3] of the Belfast group have carried out interesting experimental measurements of multiple ionization of copper by electron impact using a pulsed crossed beam technique at impact energies ranging from near thresholds to 2100 eV. The use of thermal energy Cu beams in their measurements obviates the metastable contamination inherent in the fast crossed beam method used by Freund et al. [4]. Thus, their experiment contained ground-state copper atoms ($(3d)^{10}4s$) only. They considered that at higher energies, multiple ionization is determined primarily by the Auger process following the creation of inner shell vacancies.

Among different multiple ionization processes, the double ionization is the most significant. The main contribution to the total ionization of the target is given by single- and double-ionization processes. In general, the double ionization of the target by electron impact takes place through different processes:

(i)	Direct processes	Simultaneous ejection of two electrons
(ii)	Indirect processes	Ejection of an inner shell electron followed by Auger emission
(iii)	Resonant excitation – autoionization processes.	

Rigorous theoretical calculation of the double-ionization cross-sections 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 [5]. Sophisticated calculations of the integrated double-ionization cross-sections of atoms/ions by electron impact have not been reported so far. Very recently, Belenger et al. [6] have reported a semi-empirical formula for the evaluation of double-ionization cross-sections of neutral atoms, and positive and negative ions by electron impact and also presented results for copper targets. In this approach, the shape of the cross-sections is described by analytical expressions and approximation parameters (constants) are estimated by fitting the model cross-sections to reliable experimental data. In addition to this, similar methods have been reported by Fisher et al. [7] and Deutsch et al. [8]. Keeping the above mentioned facts in view, we have thought it worthwile to apply a suitable theoretical approach, e.g. the binary-encounter method, for the calculation of double-ionization cross-sections of copper by electron impact with the aim to compare our result with the experimental observations discussed earlier. The binary-encounter theory of electron impact double ionization of atoms developed by Gryzinski [9] was modified by Roy and Rai [10]. Later on, this method was used in the case of several neutral and ionic targets with some modifications and satisfactory results were obtained [11–14]. However, the use of Hartree-Fock velocity distribution in considering the ejection of the two electrons has not been reported so far. In all previous calculations, Hartree-Fock and hydrogenic velocity distributions have been used, while considering the ejection of the first and the second target electron, respectively. The hydrogenic velocity distribution, though inappropriate, in the case of targets with more than two electrons, has been used by the previous workers for the sake of simplicity. In the present work, we made use of Hartree-Fock velocity distribution for both electrons, with the aim to achieve better accuracy.

2. Theoretical methods

According to the Gryzinski's double-binary-encounter model [9], the electron-impact double-ionization cross-section, Q^{ii}, is given by

$$Q^{ii} = Q^{ii}_{\rm sc} + Q^{ii}_{\rm ej}\,, \tag{1}$$

where $Q^{ii}_{\rm sc}$ is the cross-section for ejection of the two electrons by the incident electron and $Q^{ii}_{\rm ej}$ the cross-section for double ionization when the first ejected electron knocks out the second electron. The expressions for the two processes, as given by Gryzinski [9] and modified by Roy and Rai [10], are

$$Q^{ii}_{\rm sc} = \frac{n_e(n_e-1)}{4\pi\bar{r}^2} \int\limits_{U_i}^{E_q-U_{ii}} \sigma_{\Delta E} \left[\int\limits_{U_{ii}}^{E_q-\Delta E} \sigma_{\Delta E'}\ {\rm d}(\Delta E') \right] {\rm d}(\Delta E)\,, \tag{2}$$

and

$$Q^{ej}_{\rm sc} = \frac{n_e(n_e-1)}{4\pi\bar{r}^2} \int\limits_{U_i+U_{ii}}^{E_q} \sigma_{\Delta E} \left[\int\limits_{U_{ii}}^{\Delta E-U_i} \sigma_{\Delta E'}\ {\rm d}(\Delta E') \right] {\rm d}(\Delta E)\,. \tag{3}$$

The symbols in the above expressions have been defined by Gryzinski (see also Roy and Rai).

In order to obtain an analytic expression for $Q^{ii}_{\rm sc}$ and $Q^{ii}_{\rm ej}$, Gryzinski estimated the cross-section for the second collision for average value of the energy transfer and assumed an exponential velocity distribution for the target electron, which is not justified physically. Further, Vriens [15] found that the expression for $\sigma_{\Delta E}$ used by Gryzinski was incorrect. These shortcomings have been reported by Roy and Rai [10].

An accurate expression for $\sigma_{\Delta E}$ given by Vriens [15] in a symmetrical model, including exchange and interference, has been used in the present calculations. Following Catlow and McDowell [16], two dimensionless variables, s and t, are 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 and target electrons, respectively, and $u = v_0^2$ is the ionization potential of the target in Rydbergs. All other energies involved have also been expressed in Rydbergs. In terms of these variables, $\sigma_{\Delta E}$ is given by (see Kumar and Roy [11], Chatterjee and Roy [13])

$$\sigma_{\Delta E} \;=\; \frac{2}{s^2u+t^2u+u}\left[\frac{1}{\Delta E^2}+\frac{4t^2u}{3\Delta E^3}\right.$$
$$\left.+\frac{1}{(s^2u+u-\Delta E)^2}+\frac{4t^2u}{3(s^2u+u-\Delta E)^3}-\frac{\Phi}{\Delta E(s^2u+u-\Delta E)}\right],$$

where

$$\Phi = \cos\left(\sqrt{\frac{1}{s^2 u + u}}\;\ln s^2\right) . \tag{4}$$

The expressions for cross-sections have been integrated numerically over energy transfer and Hartree-Fock momentum distribution for ejection of the two electrons. Thus, expressions (2) and (3) take the form

$$Q^{ii}_{\rm sc} = \frac{n_e(n_e-1)}{4\pi\bar{r}^2} \int\limits_{t=0}^{\infty} \int\limits_{U_i}^{E_q-U_{ii}} \sigma_{\Delta E} \left[\int\limits_{t=0}^{\infty} \int\limits_{U_{ii}}^{E_q-\Delta E} \sigma_{\Delta E'} f(t)\sqrt{U_{ii}}\;{\rm d}(\Delta E')\;{\rm d}t \right]$$

$$\times f(t)\sqrt{U_i}\;{\rm d}(\Delta E)\,{\rm d}t \times 8.797\times 10^{-17}\,(\pi a_0^2)\,, \tag{5}$$

and

$$Q^{ii}_{\rm ej} = \frac{n_e(n_e-1)}{4\pi\bar{r}^2} \int\limits_{t=0}^{\infty} \int\limits_{U_i+U_{ii}}^{E_q} \sigma_{\Delta E} \left[\int\limits_{t=0}^{\infty} \int\limits_{U_{ii}}^{\Delta E-U_i} \sigma_{\Delta E'} f(t)\sqrt{U_{ii}}\;{\rm d}(\Delta E')\;{\rm d}t \right]$$

$$\times f(t)\sqrt{U_i}\;{\rm d}(\Delta E)\,{\rm d}t \times 8.797\times 10^{-17}\,(\pi a_0^2)\,. \tag{6}$$

There is strong experimental evidence (Bolorizadeh et al. [3]) that at higher energies Auger transitions contribute largely to the process of double ionization. In order to include this mechanism of double ionization, i.e., to compute the value of Q^{ii}_A, we have calculated electron impact single-ionization cross-sections for 3p- and 3s-shells. Q^{ii}_A is given by Chatterjee and Roy [13] as

$$Q^{ii}_A = a n_e \int\limits_0^{\infty} \left\{ \frac{4}{(s^2+t^2+1)u^2} \left[\frac{s^2-1}{s^2} + \frac{2t^2}{3}\,\frac{s^4-1}{s^4} - \frac{\Phi \ln s^2}{s^2+1} \right] \right\} \times f(t)\sqrt{u}\,{\rm d}t(\pi a_0^2)\,. \tag{7}$$

The various symbols appearing in the above equation have already been defined by Chatterjee and Roy [13].

The momentum distribution function $f(t)$ is given by (see Ref. [16])

$$f(t) = 4\pi t^2 u \rho_{\rm nl}(t\sqrt{u})\,. \tag{8}$$

Here

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

where

$$\Psi_{nlm}(\vec{x}) = \frac{1}{(2\pi)^{3/2}} \int \Phi_{nlm}(\vec{r}) \mathrm{e}^{\mathrm{i}\vec{k}\cdot\vec{r}}\, \mathrm{d}\vec{r}$$

is the Fourier transform of the one electron orbital

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

in which $R_{nl}(r)$ is the Hartree-Fock radial function.

In Eq. (5), u and s^2 have been replaced by U_i and E_q/U_i in 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 Eq. (6) is that s^2 assumes the value $(\Delta E - U_i)/U_{ii}$ in expression for $\sigma_{\Delta E'}$. $f(t)$ has been constructed by replacing u by U_i for the ejection of the first electron and by U_{ii} in case of the second electron. In the present case of copper $n_e(n_e - 1)$ has been replaced by $n_{e_1} \times n_{e_2}$ where $n_{e_1} = 1$ and $n_{e_2} = 10$ considering ejection of one electron from 4s- and the other from 3d-shell. The value of $\bar{r}$ has been obtained by dividing the mean value of the radii of these shells by the cube root of the sum of the number of electrons in the shells (see Kumar and Roy [11]). The values of Q^{ii}_{sc} and Q^{ii}_{ej} are exactly equal at all incident energies as in the Vriens treatment after the first encounter, the ejected and incident electrons are treated as indistinguishable (see Roy and Rai [10], Kumar and Roy [11]). Hence in order to obtain double-ionization cross-section, the value given by the expression for Q^{ii}_{sc} (or Q^{ii}_{ej}) should be multiplied by 2.

For binding energies of electrons, we have used the values of Hattree-Fock orbital energies of shells of Cu ($(3\mathrm{d})^{10}4\mathrm{s}$) and Cu^+ ($(3\mathrm{d})^{10}4\mathrm{s}^0$) as given by Clementi and Roetti [17] in the present calculations. The quantum mechanical values of the maximum radial probability density reported by Desclaux [18] have been used as shell radii. Momentum distribution function for the target electron $f(t)$ has been constructed using the Hartree-Fock radial wave functions taken from Clementi and Roetti [17].

Here we would like to mention that consideration of the correlation of events (dynamic correlation) as well as the electron-electron correlation is desirable in calculations of direct double ionization cross-sections (see Deb and Crothers [19]). The use of correlated wave function for the target electrons takes into account the electron-electron correlation in the theoretical studies of double-ionization process (see Griffin and Pindzola [20]). Calculations using these wave functions become very complicated particularly for heavier targets. Jha et al. [21] have discussed that study of double ionization in Gryzinski's model using Hartree-Fock wave function for the target electrons takes into account dynamic correlation as well as electron-electron correlation to some extent and hence such studies may be reasonable.

3. *Results and discussion*

We have calculated electron impact double-ionization cross-section from 35 eV to 1000 eV using the method discussed in Sect. 2. The theoretical results along with experimental data have been presented in Fig. 1 and Table 1. We have also presented results of semiempirical calculation [6] in Fig. 1 and Table 1 for the sake of comparison. In the threshold region, it is expected that double ionization will take place due to removal of one electron from the 4s- and the other from the 3d-shell. The orbital energies of 4s- and 3d-shells of neutral copper, being 6.5 and 13.3 eV, are not very close. At low incident energies, after the ejection of the first electron from the 4s-shell, the target may get sufficient time for rearrangement and thus one would expect the second electron to be ejected from the 3d-shell of Cu^+, the threshold for double-ionization being 28.5 eV. We have used orbital energies and Hartree-Fock wave functions of 4s-shell of Cu and of 3d-shell of Cu^+ ($(3d)^{10}4s^0$) in our calculations. In order to include contributions of Auger emission to double-ionization cross-sections, we have calculated electron impact single-ionization cross-sections for 3p- and 3s-shells. Keeping in view the dominant contribution due to Auger emission [3] and non availability of Auger yield in the literature, we have assumed the Auger yield to be unity. It is observed that up to 40 eV, our cross-sections are in reasonable agreement with experiment, being almost two times smaller than the mean experimental values. In the energy range 50 eV – 100 eV, the theoretical cross-sections diverge more and are about 3 – 7 times smaller than the experimental data. This discrepancy may be attributed to the contribution to double ionization from removal of two 3d-electrons which could not be included in the present calculations due to non-availability of orbital energies and

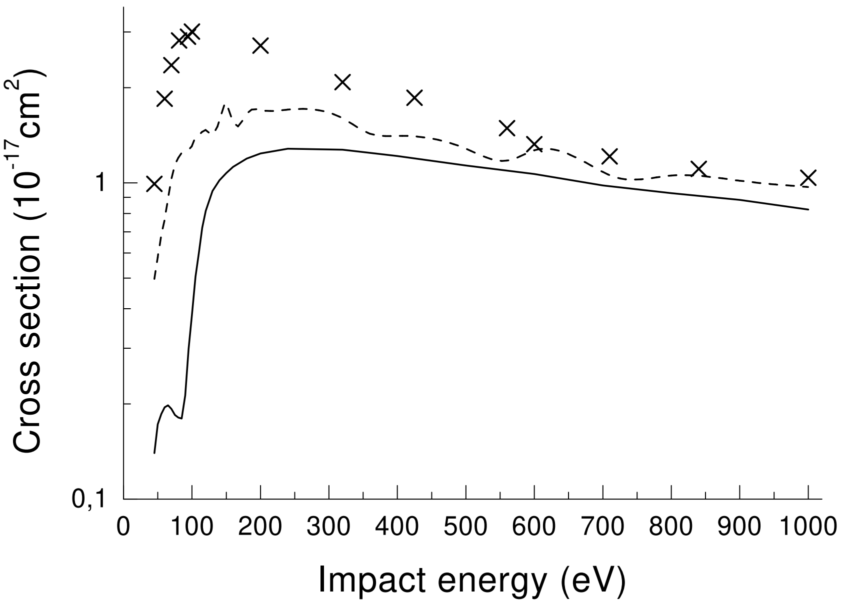

Fig. 1. Electron impact double ionization cross-sections of copper: present results (full curve), experimental data (Ref. [3], dashed curve) and semiempirical calculation (Ref. [6], crosses).

Hartree-Fock wave functions for the $(3d)^9 4s$ configuration of copper. Beyond 100 eV impact energy, the calculated cross-sections, though smaller than the experimental values, lie well within a factor of two. With increase in energy, the present results become closer to experiment, and beyond 425 eV impact energy they are only about 10 % smaller than the mean experimental cross-sections. Undoubtedly, the use of correct value of Auger yield would reduce the cross-sections leading to a larger difference between the calculated results and experimental data. This is physically reasonable as double ionization of two $n = 3$ electrons has not been included in the calculations. It can be seen that the semi-empirical calculations of Belenger et al. [6] considerably overestimate the cross-sections from near threshold up to 500 eV impact energy.

Table 1. Electron-impact double-ionization cross-sections of copper (in units of 10^{-17} cm^2).

Energy (eV)	This work				Exper. Ref. [3]	Semiemp. calc. [6]
	Direct double ionization	Auger contr.		Total		
		3p vacancy	3s vacancy			
35	0.09	–	–	0.09	0.15	–
40	0.14	–	–	0.14	0.29	1.0
50	0.18	–	–	0.18	0.59	1.9
60	0.20	–	–	0.20	0.79	2.3
70	0.19	–	–	0.19	1.05	2.8
81	0.18	–	–	0.18	1.21	2.9
94	0.17	0.12	–	0.29	1.27	–
100	0.16	0.30	–	0.46	1.37	3.0
126	0.14	0.76	–	0.90	1.40	–
159	0.11	1.00	0.05	1.16	1.54	–
200	0.09	1.12	0.09	1.30	1.69	2.7
320	0.06	1.11	0.13	1.30	1.60	2.1
425	0.04	1.04	0.13	1.21	1.35	1.9
560	0.03	0.94	0.12	1.09	1.18	1.5
600	0.03	0.91	0.12	1.06	1.28	1.3
710	0.02	0.84	0.12	0.98	1.07	1.2
840	0.02	0.77	0.11	0.90	1.01	1.1
1000	0.02	0.70	0.11	0.82	0.96	1.0

Good agreement of the present calculations with experimental data beyond 100 eV supports the view [3] that at higher energies removal of inner-shell electron starts to dominate and in that case Auger contribution would become very important. In this connection it may be noted that ionization threshold for 3p- and 3s-shells are 90.3 and 136 eV, respectively. It is seen from the table of experimental results [3] that double-ionization cross-section is not a smooth function of energy, fluctuations being observed at several energy values. However, there are two peaks of larger cross-sections, 1.78×10^{-17} cm^2 and 1.69×10^{-17} cm^2, at incident energies 147 eV and 200 eV, respectively. The present calculations do not show a peak at 147 eV but a peak of magnitude 1.31×10^{-17} cm^2 is obtained at 200 eV which is in good agreement with the experimental observations.

In the discussion of experimental results [3], the ionization of two $n = 3$ electrons has also been indicated. Considering removal of either two 3s-eletrons, one 3s-electron plus one 3p-electron or two 3p-electrons, the threshold energies for such processes have been shown to be between 210 eV and 315 eV [3]. These data suggest that ionization threshold for ejection of the second electron has been considered when the atom becomes ionized after removal of the first electron. It has not been possible to include these contributions in the present work due to the non-availability of particular ionization thresholds and Hartree-Fock wave functions corresponding to the ejection of the second electron. However, these double-ionization cros-sections are expected to be much smaller in comparison to the dominant Auger contributions. From Table 1 it is seen that at 1000 eV the difference between the present result and the experimental value is larger than that at lower impact energies. This may be attributed to the onset of Auger emission following the creation of a 2p-vacancy, the ionization threshold being 969 eV.

4. *Conclusion*

Keeping in view the complexity of the problem and the discussions given above, it is concluded that the present method using Hartree-Fock momentum distribution for ejection of two electrons gives a satisfactory account of the experimental observations.

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DVOSTRUKA IONIZACIJA BAKRA UDAROM ELEKTRONA

Istražili smo dvostruku ionizaciju bakra udarom elektrona u izmijenjenom modelu dvojnih binarnih sudara. Prvi se puta u razmatranjima izbacivanja dvaju elektrona iz mete u binarnim sudarima primjenjuje Hartree-Fockova raspodjela brzina. Doprinos Augerove emisije, koji dominira na višim udarnim energijama, također smo uključili u račun. Dobiveni se ishodi računa uspoređuju s nedavnim mjerenjima Bolorizadeha i sur. i poluempirijskim računima Belengera i sur. Nalazimo da poluempirijski računi precjenjuju udarne presjeke. Naši su ishodi računa u dobrom skladu s eksperimentalnim vrijednostima.