

## Single and double ionization of gallium by electron impact

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**Abstract.** Electron impact single and double ionization cross sections of gallium have been calculated in the binary encounter approximation using accurate expression for  $\sigma_{\Delta E}$  including exchange and interference as given by Vriens and Hartree–Fock velocity distributions for the target electrons throughout the calculations. It is concluded that the ionization of 3d shell contributes partly to single ionization and partly to double ionization. The results so obtained show reasonably good agreement with the experimental data.

**Keywords.** Electron impact; single ionization; double ionization; gallium.

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### 1. Introduction

Cross sections for ionization of atoms/ions by electron impact find wide applications in various branches of science and technology. In particular, study of electron impact multiple ionization of atoms is of great importance in understanding astrophysical and fusion plasmas. Considering the relevance of reliable data on gallium to tokamak fusion devices, latest experimental measurements of single and multiple ionization cross sections for gallium have been carried out by Patton *et al* [1] using a pulsed crossed beam technique incorporating time of flight spectroscopy.

The experimental data on single ionization have been compared with the empirical and semiclassical calculations of Lotz [2] and Margreiter *et al* [3] respectively. The double ionization cross sections have been compared only with the predictions of scaling law proposed by Fisher *et al* [4]. Fortunately the experimental measurements have been found to be in good agreement with the scaled cross sections. The calculations of single ionization cross section consider ionization of 4p and 4s shells only on the ground that the removal of a 3d electron leads to a large number of states whose energy exceeds the threshold for double ionization (see Patton *et al* [1]). Earlier, Vainshtein *et al* [5] compared their experimental measurements on gallium with the ionization cross sections calculated by different theoretical methods. They stated that the available published data were slightly contradictory and according to Kozlov [6], a large number of  $3d^9 4s^2 4p$  terms are characterized by an LS energy exceeding the double ionization energy of the atom. Consequently these terms experience pre-ionization decay and contribute to double ionization. At the same

time they expressed the view that a reliable separation of the calculated values of  $\sigma^+$  and  $\sigma^{++}$  requires careful study.

In calculations of ionization cross sections difficulties have been experienced by earlier workers in the case of several atoms and ions involving ionization from fully occupied  $d$  shells probably due to different physical processes and in such cases partial contributions from ionization of  $d$  shells have been taken into account to explain the experimental results. Bell *et al* [7] observed a large discrepancy between their calculated values in configuration averaged distorted wave (CADW) approximation and experimental data in the case of electron impact single ionization of  $\text{In}^+$ . In order to obtain satisfactory agreement with experimental data, the contribution of the electrons of  $4d$  shells to the ionization cross sections was added at only one half of its calculated value. Use of only half of the  $d$ -shell contributions was first proposed by Rogers *et al* [8] and has since been shown to fit the experimental data better for several experiments [9,10]. In the case of ions  $\text{Sn}^{+}$ – $\text{Ba}^+$ , Younger [11] surmized that ionization of the  $4d$  sub-shells can be followed by the autoionization of the residual ions resulting in double ionization. At the same time he has mentioned that this process has not yet received adequate enough study to allow meaningful extraction of its influence. All of the  $4d$  hole states may not necessarily decay by autoionization because of other possible mode of decay to stable bound configurations. In their work on electron impact single ionization of copper, Jha *et al* [12] have considered the contribution of one  $3d$  shell electron to single ionization which is based on critical reasoning. Due to non-availability of rigorous theoretical calculations including calculations of branching ratios for different physical processes in literature, it has been a prevalent practice to consider partial contribution of ionization of  $d$  shell to different degrees of ionization of atom/ion.

From a close inspection of the single ionization cross sections presented in table 1 by Patton *et al* [1] it is seen that there is a pronounced maximum of magnitude  $11.0 \times 10^{-16} \text{ cm}^2$  at 27.1 eV impact energy. This maximum can be considered to arise from contributions of ionization of  $4p$  and  $4s$  shells whose binding energies are 5.6 eV and 11.54 eV respectively. Beyond an impact energy of 32 eV, the cross sections go on decreasing up to an impact energy of 43 eV and reaches a maximum of  $10.1 \times 10^{-16} \text{ cm}^2$  at 46 eV impact energy. The threshold for ionization of  $3d$  shell is 32.45 eV and it is probable that the gradual increase of  $3d$  shell contribution produces this maximum. It is observed that beyond 46 eV impact energy the cross sections go on continuously decreasing except slight fluctuations at 465 eV and 560 eV which may be considered to be the usual feature of single ionization cross sections at higher energies. The above mentioned observations reflect some contributions of ionization of  $3d$  shell to single ionization cross sections.

At this stage we would like to recollect that single ionization cross sections have been calculated by empirical and semiclassical methods (see Patton *et al* [1]) whereas the double ionization cross sections have been determined by the scaling law proposed by Fisher *et al* [4]. To the best of our knowledge there has been no attempt to estimate the magnitudes of the contributions of ionization of  $3d$  shell to single and double ionization cross sections. Keeping in view the facts stated above it has been considered worthwhile to calculate both single and double ionization cross sections by a consistent theoretical approach in order to examine the above mentioned contributions.

Rigorous theoretical calculation of the double ionization cross sections becomes complicated as it involves the consideration of four charged particles in the final channel [13]. Sophisticated calculations of the integrated double ionization cross sections of atoms/ions

by electron impact are not available in the literature. As a consequence of this, semiempirical and semiclassical approaches have emerged for calculation of double ionization cross sections [4,14–16]. Recently Gryzinski and Kunc [17] using classical binary encounter approximation derived general analytical expressions for electron impact double ionization cross sections of atoms with atomic number  $Z \gtrsim 20$  and  $s$  or  $d$  outer shells with two electrons. They compared their calculations only with experimental data for Ca, Sr, Ba and Hg atoms. This model is consistent and convenient but it treats the process of double ionization in a ‘statistical’ way. However, this model cannot be used for gallium. Here we would like to point out that the wave functions representing the bound electrons are the characteristics of the target atom but there is no consideration of wave functions in the above mentioned calculations. As regards electron impact single ionization, there have been a good number of quantum calculations for various atoms and ions but unfortunately in the case of gallium no such calculation is available in the literature.

Keeping in view the above mentioned facts it has been considered desirable to apply the binary encounter approximation (BEA) using Hartree–Fock velocity distribution for the target electron for calculation of single and double ionization cross sections of gallium. In the past the BEA has been found successful in the calculations of electron impact single ionization cross sections of atoms [18,19]. The double binary encounter model of electron impact double ionization developed by Gryzinski [20] was modified by Roy and Rai [21]. Later on this model with some modifications was used in the case of several atomic and ionic targets and satisfactory results were obtained [22–24]. In these calculations Hartree–Fock and hydrogenic velocity distributions were used while considering the ejection of the first and the second target electrons respectively. Gryzinski and Kunc [17] have given a discussion on the works of Roy and co-workers [22,24] in detail and pointed out their strengths and weaknesses. They have appreciated the consideration of the contributions of the  $p$  electrons from the next inner shell to double ionization of Sr and Ba atoms, prediction of acceptable magnitudes of the cross section of double ionization and the appearance of the secondary peak in the cross section. As regards the weakness of the calculation, they have expressed the view that the use of hydrogenic velocity distribution while considering the ejection of the second electron, particularly from the inner shell is physically not justified. Jha and Roy [25] made an improvement over earlier calculations by using Hartree–Fock velocity distribution for both electrons in the calculation of electron impact double ionization of copper. In the present work we have adopted the same approach for calculation of direct double ionization cross sections. In this context it may be noted that calculations using correlated wave functions become very complicated particularly for heavier targets. Jha *et al* [24] have discussed that the study of double ionization in Gryzinski’s model using Hartree–Fock wave functions for target electrons takes into account dynamic correlation as well as electron–electron correlation to some extent and hence such studies may be reasonable.

## 2. Theoretical methods

Vriens expression [26] in symmetrical model including exchange and interference has been used for calculating electron impact single ionization cross sections. The expression used in the calculation has been discussed in detail by Roy and Rai [18] and Jha *et al* [12]. A brief outline of the method of calculation is given below. Using dimensionless variables

introduced by Catlow and McDowell [27], the expression for cross section for a particular incident energy and a particular velocity of the bound electron can be written in the form

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

where

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

Numerical integration of the expression for  $Q^i(s, t)$  has been carried out over Hartree–Fock velocity distribution 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

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

The method of calculating electron impact double ionization cross sections of atoms in double binary encounter model has been discussed in detail in earlier publications [21,25,28]. However, a brief account of the expressions used in the calculations seems to be desirable. Electron impact double ionization cross section of gallium, including the contribution from the Auger emission can be written as

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

where  $Q_D^{ii}$  denotes the contribution from direct ejection of the two electrons including contributions from inner shells and  $Q_A^{ii}$  that from the 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

$$Q_{sc}^{ii} = \frac{n_e(n_e - 1)}{4\pi\bar{r}^2} \int_{t=0}^\infty \int_{U_i}^{E_q - U_{ii}} \sigma_{\Delta E} \left[ \int_{t=0}^\infty \int_{U_{ii}}^{E_q - \Delta E} \sigma_{\Delta E'} f(t) U_{ii}^{1/2} d(\Delta E') dt \right] \times f(t) U_i^{1/2} d(\Delta E) dt \times 8.797 \times 10^{-17} (\pi a_0^2) \quad (4)$$

and

$$Q_{ej}^{ii} = \frac{n_e(n_e - 1)}{4\pi\bar{r}^2} \int_{t=0}^\infty \int_{U_i + U_{ii}}^{E_q} \sigma_{\Delta E} \left[ \int_{t=0}^\infty \int_{U_{ii}}^{\Delta E - U_i} \sigma_{\Delta E'} f(t) U_{ii}^{1/2} d(\Delta E') dt \right] \times f(t) U_i^{1/2} d(\Delta E) dt \times 8.797 \times 10^{-17} (\pi a_0^2). \quad (5)$$

In the present work we have used the accurate expression for  $\sigma_{\Delta E}$  including exchange and interference as given by Vriens [26]. Using dimensionless variables introduced by Catlow and McDowell [27]  $\sigma_{\Delta E}$  is given by (see [28])

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

where

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

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.

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

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

where  $Q_D^{ii}(4p, 4s)$  stands for the double ionization cross section corresponding to the one electron ejected from the  $4p$  shell and the other from the  $4s$  shell. The factor  $n_e(n_e - 1)/4\pi\bar{r}^2$  has been suitably modified for considering the first mode of ionization. In case of gallium  $n_e(n_e - 1)$  has been replaced by  $n_{e1} \times n_{e2}$  where  $n_{e1} = 1$  and  $n_{e2} = 2$ . In order to obtain the value of  $\bar{r}$ , the atomic radius has been replaced by the mean of the expectation values of radii of the shells. We have used orbital energies of the shells of Ga and  $Ga^+$  as given by Clementi and Roetti [29] in the present calculations. The expectation values of radii reported by Desclaux [30] have been used as shell radii. Momentum distribution functions for the target electrons have been constructed using Hartree–Fock radial wave functions given by Clementi and Roetti [29].

### 3. Results and discussion

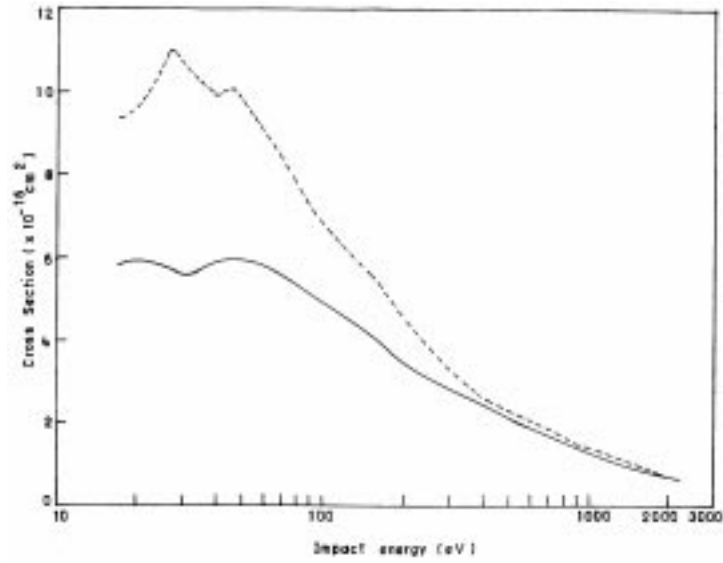
In the present work attempt has been made to obtain single and double ionization cross sections by estimating the contributions of ionization of  $3d$  shell to these cross sections. Before discussing the results we would like to mention that we have considered the contribution of eight electrons of  $3d$  shell to single ionization and those of the remaining two electrons to double ionization, the reasons for which would be given below. For this purpose we have calculated single ionization cross sections for  $4p$ ,  $4s$  and  $3d$  shells. At the same time direct double ionization cross sections have been calculated corresponding to ejection of  $(4p, 4s)$  and  $(4s, 4s)$  electrons. These calculations have been performed from 17.8 eV to 2100 eV impact energy using the method given in §2. The results of single ionization cross sections along with experimental data have been presented in figure 1. The contributions of  $4p$ ,  $4s$  and eight electrons of  $3d$  shell to single ionization cross sections have been shown separately in table 1 for the sake of critical comparison with the experimental results. First we would like to discuss our results by considering ionization of  $4p$  and  $4s$  shells only. It is seen that the calculated results differ from the experimental data within a factor of two up to 40 eV impact energy. In the energy range 43–200 eV the calculated results differ by a factor more than two from the experimental results. Further, it is observed that in the energy region 200–600 eV the theoretical and experimental results differ by a factor 2.5. Beyond 600 eV the discrepancy increases further and the two results differ by a factor more than three. In this connection it may be noted that calculations of single ionization cross sections in the BEA using Hartree–Fock velocity distribution for the target electron show good agreement with experimental data in high energy region being always

**Table 1.** Electron impact single ionization cross sections of Ga in units of  $10^{-16}$  cm<sup>2</sup>.

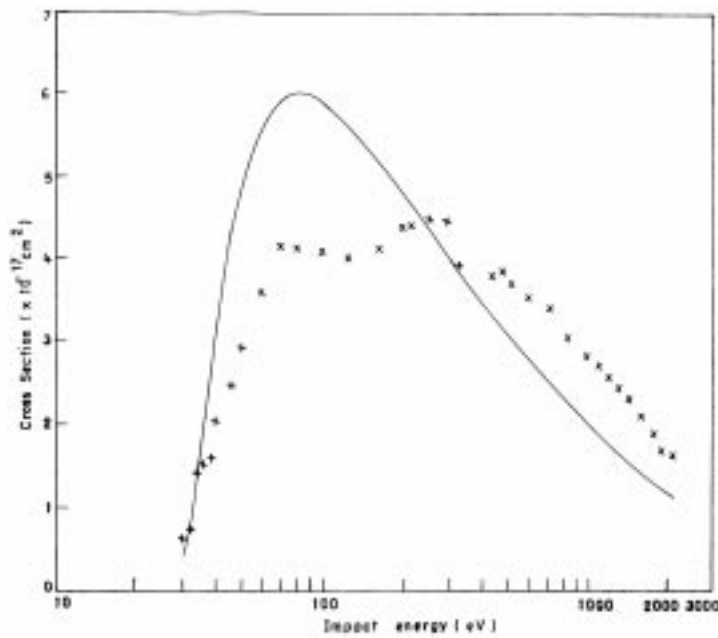
Energy (eV)	Contribution of 4 <i>p</i> shell	Contribution of 4 <i>s</i> shell	Sum of 4 <i>p</i> and 4 <i>s</i> contribution	Contribution of eight electrons of 3 <i>d</i> shell	Total	Experiment [1]
17.8	4.52	1.27	5.79		5.79	9.36
21.1	4.40	1.50	5.90		5.90	9.76
27.1	4.07	1.68	5.75		5.75	11.00
30.0	3.90	1.71	5.61		5.61	10.70
40.0	3.38	1.70	5.08	0.85	5.93	9.92
43.0	3.24	1.68	4.92	1.06	5.98	9.89
46.0	3.12	1.65	4.77	1.23	6.00	10.10
60.0	2.63	1.52	4.15	1.68	5.83	9.07
70.0	2.37	1.42	3.79	1.82	5.61	8.49
75.0	2.25	1.37	3.62	1.86	5.48	8.16
88.0	2.00	1.27	3.27	1.93	5.20	7.28
100.0	1.82	1.18	3.00	1.94	4.94	6.83
126.0	1.52	1.02	2.54	1.92	4.46	6.07
159.0	1.26	0.88	2.14	1.85	3.99	5.44
200.0	1.04	0.75	1.79	1.74	3.53	4.52
250.0	0.86	0.63	1.49	1.62	3.11	3.89
320.0	0.70	0.52	1.22	1.47	2.69	3.10
350.0	0.65	0.49	1.14	1.42	2.56	2.97
425.0	0.54	0.42	0.96	1.30	2.26	2.51
510.0	0.46	0.36	0.82	1.18	2.00	2.26
600.0	0.40	0.31	0.71	1.08	1.79	2.10
710.0	0.34	0.27	0.61	0.98	1.59	1.87
840.0	0.29	0.23	0.52	0.88	1.40	1.59
1000.0	0.25	0.20	0.45	0.78	1.23	1.48
1100.0	0.23	0.18	0.41	0.74	1.15	1.33
1200.0	0.21	0.17	0.38	0.70	1.08	1.28
1320.0	0.19	0.16	0.35	0.65	1.00	1.13
1450.0	0.17	0.14	0.31	0.61	0.92	1.10
1600.0	0.16	0.13	0.29	0.57	0.86	0.95
1775.0	0.14	0.12	0.26	0.52	0.78	0.80
1900.0	0.13	0.11	0.24	0.50	0.74	0.77
2100.0	0.12	0.10	0.22	0.47	0.69	0.70

within a factor of two. The discrepancy in the calculated cross sections considering ionization of 4*p* and 4*s* shells, and appearance of maximum at 46 eV in the experimental data lead us to believe that the ionization of 3*d* shell contributes partially to single ionization cross sections.

Now we would like to examine the calculations of double ionization cross sections. The results of double ionization cross sections along with experimental observations have been shown in figure 2 and table 2. The contributions of direct double ionization from ejection of (4*p*, 4*s*) and (4*s*, 4*s*) electrons have been shown separately in table 2. We have also presented contributions of two 3*d* electrons in a separate column in order to show contribution of Auger emission. It can be seen from table 2 that in the energy region of direct double ionization (30 and 32 eV impact energy) the calculated values agree well with experimental data. However, our results based on direct double ionization differ from the experiment



**Figure 1.** Electron impact single ionization cross sections of gallium in units of  $10^{-16}$   $\text{cm}^2$ . Solid line curve (—) and dashed line curve (- -) represent present results and experimental data [1] respectively.



**Figure 2.** Electron impact double ionization cross sections of gallium in units of  $10^{-17}$   $\text{cm}^2$ . Solid line curve (—) and multiplication sign curve ( $\times \times \times$ ) represent present results and experimental data [1] respectively.

**Table 2.** Electron impact double ionization cross sections of Ga in units of  $10^{-17}$  cm<sup>2</sup>.

Direct double ionization cross sections						
Energy (eV)	Contribution of (4 <i>p</i> , 4 <i>s</i> )	Contribution of (4 <i>s</i> , 4 <i>s</i> )	Sum of direct D.I. cross sections	Contribution of two electrons of 3 <i>d</i> shell	Total	Experiment [1]
30.0	0.45		0.45		0.45	0.63
32.0	0.62	0.01	0.63		0.63	0.73
38.0	0.96	0.13	1.09	1.70	2.79	1.59
40.0	1.02	0.17	1.19	2.12	3.31	2.05
46.0	1.12	0.25	1.37	3.08	4.45	2.48
50.0	1.14	0.29	1.43	3.52	4.95	2.95
60.0	1.09	0.33	1.42	4.20	5.62	3.62
70.0	1.01	0.34	1.35	4.56	5.91	4.17
81.0	0.90	0.33	1.23	4.76	5.99	4.16
100.0	0.73	0.29	1.02	4.86	5.88	4.09
126.0	0.56	0.24	0.80	4.80	5.60	4.04
159.0	0.42	0.19	0.61	4.62	5.23	4.15
200.0	0.31	0.14	0.45	4.36	4.81	4.42
216.0	0.28	0.13	0.41	4.26	4.67	4.45
250.0	0.22	0.11	0.33	4.06	4.39	4.50
290.0	0.18	0.09	0.27	3.84	4.11	4.47
320.0	0.15	0.08	0.23	3.68	3.91	3.95
425.0	0.10	0.05	0.15	3.24	3.39	3.86
465.0	0.09	0.05	0.14	3.10	3.24	3.90
510.0	0.08	0.04	0.12	2.94	3.06	3.75
600.0	0.06	0.03	0.09	2.70	2.79	3.57
710.0	0.05	0.03	0.08	2.44	2.52	3.47
840.0	0.04	0.02	0.06	2.20	2.26	3.07
1000.0	0.03	0.02	0.05	1.96	2.01	2.86
1100.0	0.02	0.02	0.04	1.84	1.88	2.73
1200.0	0.02	0.01	0.03	1.74	1.77	2.59
1320.0	0.02	0.01	0.03	1.62	1.65	2.45
1450.0	0.02	0.01	0.03	1.52	1.55	2.33
1600.0	0.01	0.01	0.02	1.42	1.44	2.14
1775.0	0.01	0.01	0.02	1.30	1.32	1.89
1900.0	0.01	0.01	0.02	1.24	1.26	1.71
2100.0	0.01	0.01	0.02	1.16	1.18	1.66

within a factor of 2 up to 46 eV impact energy. Beyond this energy the discrepancy goes on increasing and it is found that at energies 70, 100 and 126 eV, the direct double ionization cross sections become less than one-third, one-fourth and one-fifth of the experimental values respectively. Above 126 eV impact energy this trend becomes faster and at much higher energies the calculated results are found to be insignificant as compared to experimental data. These observations confirm that the process of direct double ionization cannot explain the experimental data and reflect partial contribution of ionization of 3*d* shell to double ionization cross sections. In order to include the contribution of the ionization of 3*d* shell, we have assumed the Auger yield to be unity due to non-availability of the same in the literature. The consideration of the contributions of ten electrons of 3*d* shell to double ionization cross sections adversely affects the results. In the energy range 38–200 eV the cross sections become much larger than the experimental data differing by a factor more



than 5. Again from 216 to 465 eV the cross sections are more than four times larger than the experimental values. Beyond 510 eV impact energy the calculated cross sections and experimental data differ by a factor between 3 and 4.

The discussions about single and double ionization cross sections given above clearly suggest that the ionization of  $3d$  shell contributes partially to single ionization and partially to double ionization. On the basis of the observations and approaches adopted by earlier workers regarding ionization of fully occupied  $d$  shell as discussed in §1, we have attempted to consider the contribution of  $3d$  shell of gallium to single and double ionization cross sections. A close inspection of the calculated results reveals the possibility that eight electrons of  $3d$  shell contribute to single ionization and remaining two electrons to double ionization. The single ionization cross sections obtained from the above mentioned consideration exhibit good agreement with experimental data. It can be seen from table 1 that our cross sections differ from the experimental data within a factor of 2 up to 70 eV impact energy. Beyond this energy the cross sections show better agreement with experiment and differ by a factor less than 1.5. This improving trend leads to excellent agreement at all energies above 250 eV. Fortunately, in the energy region above the  $3d$  shell ionization threshold, our cross sections and experimental data show a peak at the same impact energy 46 eV, the magnitudes being  $6.0 \times 10^{-16} \text{ cm}^2$  and  $10.1 \times 10^{-16} \text{ cm}^2$  respectively. At lower energy (below the  $3d$  shell ionization threshold) the present calculations and experimental observations exhibit peaks of magnitude  $5.9 \times 10^{-16} \text{ cm}^2$  and  $11.0 \times 10^{-16} \text{ cm}^2$  at 21.1 eV and 27.1 eV respectively. From the consideration of contribution of two electrons of  $3d$  shell to double ionization cross section, the magnitudes of the double ionization cross sections show good agreement with experiment differing within a factor of two throughout the energy range investigated. However, the experimental data exhibit a good number of fluctuations in the magnitudes of double ionization cross sections in the energy range 70–560 eV which cannot be explained on the basis of the present calculations. Thus we find that the experimental data are explained satisfactorily on the basis of our assumption regarding contributions of ionization of  $3d$  shell to single and double ionization cross sections.

In view of the discussion given above, the idea that ionization of  $3d$  shell contributes only to double ionization is not tenable. Our investigation has been successful in presenting qualitative features of contribution of  $3d$  shell to single and double ionization cross sections. At the same time this effort has enabled us to obtain reasonable values of electron impact single and double ionization cross sections of gallium by a consistent theoretical approach. More elaborate theoretical work is required for quantitative understanding of the problem.

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