Electron impact single and double ionization of argon

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Abstract. Electron impact single and double ionization cross-sections for argon have been calculated in the binary encounter approximation (BEA) in the energy regions ranging from respective near thresholds to 5300 eV. The accurate expression for $\sigma_{\Delta E}$ (cross-section for energy transfer ΔE) including exchange and interference as given by Vriens and Hartee-Fock velocity distributions for the target electrons have been used throughout the calculations. It is observed that consideration of ionization of 3p and 3s shells explains overall satisfactory agreement of single ionization cross-sections with the experimental data. It is also concluded that the present method gives reasonable values of direct double ionization cross-sections. Inclusion of contributions of Auger effect due to vacancy in 2p shell brings the theoretical results of double ionization cross-sections in reasonably good agreement with the experimental observations.

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1 Introduction

Collisions involving electrons with atoms and molecules are one of the most fundamental interactions in atomic and molecular physics. The ionization of the rare gas atoms by electron impact is an important process in collision physics from a basic viewpoint. These atoms have a singlet ${}^{1}S_{0}$ electronic ground state as a result of np^{6} electron configuration in the outermost electronic sub-shell for Ne, Ar, Kr and Xe, and $1s^2$ configuration for He. As a consequence of the completely filled outermost sub-shells, rare gas atoms have high ionization energies ranging from 12.13 eV for single ionization of Xe to 21.56 eV for single ionization of Ne. The simplest rare gas atom helium has ionization energy 24.54 eV. The electron impact ionization of rare gases plays a fundamental role in planetary atmospheres, pulsed power switching, gaseous dielectrics and plasma physics [1,2]. Although the cross-sections for multiple ionization of an atom are much smaller than the corresponding single ionization cross-sections and decrease rapidly with increasing stage of ionization, multiple ionization processes are important in fusion plasmas [1,2] and in all gaseous environments with an abundance of energetic electrons [3].

Since the early cross-section measurements in the 1930s, total and partial ionization cross-sections of rare gas atoms by electron impact have been extensively investigated by various groups with different types of experimental measurements. Several review articles have ap-

peared in connection with these measurements. The crosssections for single ionization now agree within 5-10%but multiple ionization cross-sections of previous measurements exhibit large discrepancies particularly at low impact energies [4]. Among different experimental measurements on rare gases, McCallion et al. [5] of the Belfast group have carried out an important work on electron impact multiple ionization of argon using a pulsed crossed beam technique incorporating time of flight spectroscopy. Cross-sections σ_n for the production of n = 1-5 times ionized argon have been determined for impact energies ranging from respective thresholds to 5300 eV. Single ionization cross-sections have been compared with different theoretical calculations in limited energy ranges but the double and higher multiple ionization cross-sections could not be compared with theoretical results due to nonavailability of the same in literature.

McCallion et al. [5] have compared $R = \sigma_2/\sigma_1$ (ratio of double to single ionization cross-section) with other measurements. The energy dependence of R has been found to be very similar to that observed for a helium target in that, at high impact energies, R approaches an energy invariant value. The high energy trend of R for argon as stated above reflects the possibility of some other physical processes contributing to double ionization. In study of Ar L-shell ionization, Langenberg et al. [6] have shown considerable total cross-section for the production of L₂₃-shell Auger electrons by electron impact. Later on Shah et al. [7] have also discussed the importance of Auger effect in multiple ionization of iron and argon. We will bring out the

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idea of indirect physical process e.g. Auger effect contributing to double ionization of argon more clearly in discussion of theoretical results obtained in the present work.

Due to extreme complexities there have been no attempts for theoretical calculations of triple and higher multiple ionization cross-sections. Electron impact integrated double ionization cross-sections of atoms and ions in the Born approximation have been reported for a few light targets e.g. H^- , He and Li⁺ (see Tweed [8,9] and McGuire [10]). Rigorous theoretical calculation of double ionization cross-section becomes extremely difficult as it is related with a 4-body Coulomb problem in the final channel [11] and hence such calculations are not available in the literature. Due to this reason semi-empirical formulae and scaling laws have been developed for calculations of multiple ionization cross-sections.

Theoretical calculations of electron impact double ionization cross-sections are considered to be of much significance because contributions from different mechanisms e.g. simultaneous ejection of two electrons, inner shell ionization followed by Auger emission, resonant excitationautoionization process etc. can be separately estimated at different impact energies. Keeping in view the difficulties in carrying out sophisticated calculations, the binary encounter approximation can be considered to provide suitable theoretical description of double ionization process. In the past the binary encounter approximation (BEA) has been used successfully to calculate electron impact single and double ionization cross-sections for several atoms and ions. In spite of certain unrealistic features and unjustified simplifications in Gryzinski's [12] mathematical formulation for the process of double ionization, the idea of the two double binary encounter processes has physical justification (see Roy and Rai [13]). These processes in fact correspond to the existence of correlation between electrons of an atom and to the finite probability of the second Born Process (see Vriens [14]). Soon after the publication of Gryzinski's theory, Vriens [15] detected errors in the work and obtained accurate expression for $\sigma_{\Delta E}$ (crosssection for energy transfer ΔE) which is used frequently in calculations of single and double ionization cross-sections. Roy and Rai [13] modified the mathematical framework of Gryzinski's theory of electron impact double ionization suitably incorporating the necessary corrections. Later on this modified model for calculation of direct double ionization cross-section was used in case of several atomic/ionic targets including contributions to double ionization from indirect physical processes [16,17] and encouraging results were obtained. In these calculations Hartree-Fock and hydrogenic velocity distributions were used while considering the ejection of the first and the second target electrons respectively.

In recent past a remarkable modification has been introduced in the method of calculation of double ionization cross-sections. This involves the use of HF velocity distribution while considering the ejection of both electrons of the target. Electron impact single and double ionization cross-sections for Mg and Pb calculated in the

binary encounter model (see Jha and Roy [18,19]) show good agreement with experimental data. Inclusion of the contributions of Auger effect to double ionization crosssections has been theoretically substantiated by these calculations. Jha [20] calculated electron impact double ionization cross-sections of C⁺, N⁺, O⁺ and Ne⁺ including the contributions of ionization-autoionization. These results show satisfactory agreement with experimental data except at low incident energies. Very recently Jha and Roy have reported electron impact double ionization crosssections for Ti ions [21] and Fe ions [22] which show satisfactory agreement with experimental observations. In these calculations the expressions of cross-sections were numerically integrated over energy transfer and HF velocity distribution for the target electrons. Encouraged by the success achieved by the above mentioned method we have considered it worthwhile to take up calculations of single and double ionization cross-sections for Ar in the BEA in order to compare the theoretical results with experimental data. At the same time this work will enable us to analyse the direct double ionization cross-sections and to identify the contributions to double ionization from Auger effect resulting from single ionization of an inner shell.

2 Theoretical methods

The theoretical methods for calculating electron impact single and double ionization cross-sections of atoms have been described by Jha and Roy [18,19]. Keeping in view the convenience for the reader we consider it worthwhile to discuss briefly the ideas and methods of calculation used in the present work.

Vriens expression [15] in symmetrical model including exchange and interference has been used for calculating electron impact single ionization cross-sections. Using dimensionless variables introduced by Catlow and McDowell [23], the expression of cross-section for a particular incident energy and a particular velocity of the bound electron can be written in the form

$$Q^{i}(s,t) = \frac{4}{(s^{2}+t^{2}+1)u^{2}} \times \left[\frac{s^{2}-1}{s^{2}} + \frac{2t^{2}}{3}\left(\frac{s^{4}-1}{s^{4}}\right) - \frac{\phi \ln s^{2}}{(s^{2}+1)}\right] (\pi a_{0}^{2}) \quad (1)$$

where

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

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

$$Q^{i}(s) = n_{e} \int_{0}^{\infty} Q^{i}(s,t) f(t) u^{\frac{1}{2}} dt.$$
 (2)

Electron impact double ionization cross-section of argon including the contribution from the 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 ejection of the two electrons including contributions from inner shell 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

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

and

W

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

In the present work we have used accurate expression for $\sigma_{\Delta E}$ including exchange and interference as given by Vriens [15]. Using dimensionless variables introduced by Catlow and McDowell [23] $\sigma_{\Delta E}$ is given by

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

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.

Due to indistinguishability of electrons in the symmetrical model of Vriens the cross-sections corresponding to the two processes are exactly equal at all incident energies (see Kumar and Roy [24]) and hence in order to obtain the direct double ionization cross-section, either of the crosssections should be multiplied by two. The function f(t)appearing in equations (2), (4) and (5) is the momentum distribution function (see Catlow and McDowell [23], Jha and Roy [18]). In order to obtain Q_A^{ii} (contribution to 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 argon as given by

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

where $Q_D^{ii}(3p, 3s)$ stands for the double ionization crosssection corresponding to the one electron ejected from the 3p shell and the other from the 3s shell. We have used the binding energies of the shells as given by Clementi and Roetti [25]. The shell radii reported by Fraga et al. [26] have been used in the present calculations. Momentum distribution functions for the target electrons have been constructed using Hartree-Fock radial wave functions given by Clementi and Roetti [25].

3 Results and discussion

First of all we would like to discuss the variation of $R = \sigma_2/\sigma_1$ (ratio of double to single ionization crosssection) with energy as presented by McCallion et al. [5]. In the energy region 50–100 eV, increasing R shows that percentage increase of double ionization cross-section in a given energy interval is faster as compared to single ionization cross-section. After attaining the maximum value, R decreases in the region 100–1000 eV. This decreasing trend signifies that percentage decrease of double ionization cross-section in a given energy interval is faster as compared to single ionization cross-section. Beyond 1000 eV it is found that R is almost energy invariant indicating similar decrease of double and single ionization cross-sections. This shows the possibility of the contributions to double ionization from indirect physical processes. Keeping the above mentioned facts and the ideas presented in the Introduction in view, it is natural to consider the possibility of ionization of L-shell contributing to double ionization through Auger effect.

Now we would consider the degree of agreement of the previous theoretical calculations of single ionization crosssections with the experimental data as presented by Mc-Callion et al. [5]. All the calculations have been carried out in different limited energy ranges and it is seen that they overestimate the cross-sections in the region of and beyond the cross-section peak values. The values calculated by Peach [27] using Ochkur approximation in the energy region 18–2000 eV are found to be in reasonable agreement with experiment only at low impact energies below the cross-section peak. The cross-sections calculated by McGuire [28] using the generalized oscillator strength approximation in the energy region 20-400 eV although larger are found to be approaching the experimental values at energies beyond the experimental cross-section peak. The calculated values based on the distorted wave approximation by Younger [29] in the energy region 25–110 eV approximately are about 1.6 times larger than experimental data in the cross-section peak region. The calculation

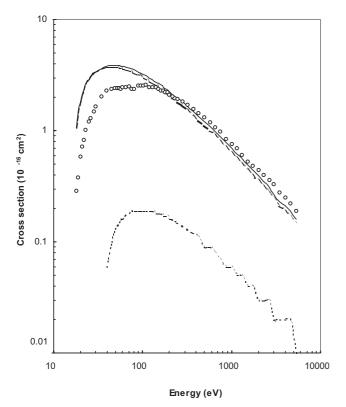


Fig. 1. Electron impact single ionization cross-section of argon: (- - -) contributions of 3p shell; (- - - -) contributions of 3s shell; (---) total; $(\circ \circ \circ)$ experimental data [5].

by Bartschat and Burke [30] using the R-matrix approach in the energy region 20–110 eV exhibits the best overall agreement with experiment. In the cross-section peak region these values are about 1.4 times the experimental data.

We have calculated electron impact single ionization cross-sections for 3p, 3s, 2p and 2s shells of argon atom and found that ionization cross-sections for 2s shell are negligible. In case of direct double ionization, contributions from inner shell have also been included in the calculations. It is found that contribution to direct double ionization from ejection of (3p, 2p) electrons is insignificant. We have attempted to analyse the single and direct double ionization cross-sections and to identify the inner shell whose ionization leads to Auger effect contributing to double ionization.

Our calculated results of single ionization cross-section considering the contributions of 3p and 3s shells only along with experimental data in the energy range 18–5300 eV have been presented in Figure 1 and Table 1. It is found that ionization cross-sections of 3s shell are much smaller than those of 3p shell. 2p ionization cross-sections (shown in Tab. 2) have not been included because they produce insignificant change in single ionization cross-sections. As the previous theoretical calculations have been carried out in different limited energy ranges and discussed earlier, we have not presented them in the table and the figure. The present results of single ionization overestimate the cross-

Table 1. Electron impact single ionization cross-sections of Ar	
in units of 10^{-16} cm ² .	

Energy	Contributions	Contributions		Experiment
(eV)	of $3p$ shell	of $3s$ shell	Total	[5]
18.0	1.06		1.06	0.29
20.0	1.81		1.81	0.58
23.0	2.65		2.65	1.02
25.0	2.89		2.89	1.20
26.5	3.08		3.08	1.30
28.0	3.28		3.28	1.49
34.5	3.61		3.61	2.05
49.5	3.73	0.13	3.86	2.40
54.5	3.69	0.15	3.84	2.40
60.0	3.62	0.16	3.78	2.46
70.0	3.48	0.18	3.66	2.49
80.0	3.33	0.19	3.52	2.39
90.0	3.18	0.19	3.37	2.55
108.0	2.94	0.19	3.13	2.57
120.0	2.79	0.19	2.98	2.48
130.0	2.68	0.19	2.87	2.47
140.0	2.57	0.18	2.75	2.44
160.0	2.39	0.18	2.57	2.30
180.0	2.22	0.17	2.39	2.23
200.0	2.09	0.16	2.25	2.10
220.0	1.96	0.16	2.12	2.05
250.0	1.80	0.15	1.95	1.94
280.0	1.68	0.14	1.82	1.83
375.0	1.37	0.12	1.49	1.56
430.0	1.24	0.11	1.35	1.45
500.0	1.11	0.09	1.20	1.31
570.0	1.00	0.09	1.09	1.19
650.0	0.91	0.08	0.99	1.07
750.0	0.81	0.07	0.88	0.98
870.0	0.72	0.06	0.78	0.84
1000.0	0.64	0.06	0.70	0.76
1150.0	0.57	0.05	0.62	0.69
1320.0	0.51	0.05	0.56	0.61
1520.0	0.45	0.04	0.49	0.53
1750.0	0.40	0.04	0.44	0.48
2000.0	0.36	0.03	0.39	0.44
2300.0	0.32	0.03	0.35	0.40
2650.0	0.28	0.03	0.31	0.36
3000.0	0.25	0.02	0.27	0.33
3500.0	0.21	0.02	0.23	0.28
4000.0	0.19	0.02	0.21	0.25
4600.0	0.17	0.02	0.19	0.22
5300.0	0.15	0.01	0.16	0.20

sections in the energy region close to threshold but become within a factor of 2 of experimental data beyond 34.5 eV. The calculated values go on improving with increase in energy and are found to be within a factor of 1.5 beyond 70 eV. The agreement of our results is found to be better and better with increasing energy and the values become within a factor of 1.25 of the experimental results beyond 108 eV. The gradually improving trend continues at higher energies also and it is remarkable that the theoretical results are almost identical to the experimental values in the energy region 250–280 eV. Beyond this energy region the experimental cross-sections become larger than the calculated ones but they always remain within a factor of 1.25 up to the highest energy considered (5300 eV).

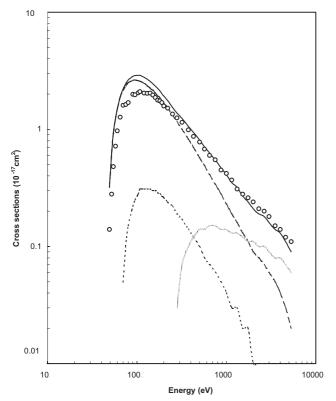


Fig. 2. Electron impact double ionization cross-section of argon: (- - -) contributions of (3p, 3p); (- - - -) contributions of (3p, 3s); (....) contributions of Auger effect due to 2p shell; (--) total; $(\circ \circ \circ)$ experimental data [5].

The double peak structure in the experimental crosssections at 90 eV and 108 eV has been ascribed to an auto ionization process involving the 3d and 4p levels of argon (see McCallion et al. [5]). Unfortunately the calculation of the autoionization process is not feasible in the BEA. However, our calculated cross-sections are about 1.3 times and 1.2 times the magnitudes of the experimental peaks at 90 eV and 108 eV respectively. The peak in our calculations at 49.3 eV is shifted considerably on the left side probably due to overestimation of cross-sections at low impact energies. A critical comparison of our results with previous theoretical calculations reveals that overestimation of the present cross-sections in the experimental peak region is less pronounced. Keeping the above mentioned discussion of the previous calculations and our theoretical results in view it is concluded that the present calculations of single ionization cross-sections show overall satisfactory agreement with the experiment in the entire energy region investigated.

The theoretical results of double ionization crosssections along with the experimental data have been presented in Figure 2 and Table 2. The calculated results of double ionization cross-sections from ejection of (3p, 3p), (3p, 3s) electrons have been shown separately. It is seen that the direct double ionization cross-sections based on the above mentioned contributions are much smaller as compared to the experimental values at high impact energies. Keeping this discrepancy in view, the contributions

Table 2. Electron impact double ionization cross-sections of Ar in units of 10^{-17} cm².

Ar in un	Contri-	Contri-	Contributions		Exper-
Energy	butions	butions	of $2p$ single		iment
(eV)	of $(3p, 3p)$	of $(3p, 3s)$	ionization	Total	[5]
49.3	0.32			0.32	0.14
52.0	0.63			0.63	0.28
54.5	0.92			0.92	0.48
60.0	1.49			1.49	0.97
65.0	1.88			1.88	1.27
75.0	2.36	0.10		2.46	1.63
80.0	2.49	0.15		2.64	1.69
90.0	2.62	0.22		2.84	1.99
95.0	2.63	0.25		2.88	1.97
108.0	2.58	0.31		2.89	2.10
120.0	2.48	0.31		2.79	2.04
130.0	2.37	0.31		2.68	2.03
140.0	2.26	0.31		2.57	2.04
150.0	2.15	0.30		2.45	1.96
160.0	2.04	0.30		2.34	1.87
170.0	1.94	0.29		2.23	1.78
180.0	1.85	0.28		2.13	1.75
200.0	1.67	0.26		1.93	1.58
220.0	1.52	0.24		1.76	1.52
250.0	1.32	0.22		1.54	1.35
280.0	1.16	0.19	0.03	1.38	1.25
320.0	1.00	0.17	0.07	1.24	1.15
375.0	0.82	0.14	0.10	1.06	0.99
500.0	0.57	0.10	0.14	0.81	0.78
570.0	0.49	0.08	0.14	0.71	0.68
650.0	0.41	0.07	0.15	0.63	0.60
750.0	0.34	0.06	0.15	0.55	0.55
870.0	0.28	0.05	0.14	0.47	0.45
1000.0	0.23	0.04	0.14	0.41	0.42
1150.0	0.19	0.03	0.13	0.35	0.37
1320.0	0.16	0.03	0.13	0.32	0.31
1520.0	0.13	0.02	0.12	0.27	0.28
1750.0	0.11	0.02	0.11	0.24	0.26
2000.0	0.09	0.01	0.11	0.21	0.24
2300.0	0.08	0.01	0.10	0.19	0.22
2650.0	0.07	0.01	0.10	0.18	0.20
3000.0	0.06	0.01	0.09	0.16	0.18
3500.0	0.05	0.01	0.08	0.14	0.15
4000.0	0.04	0.01	0.08	0.13	0.14
4600.0	0.03	0.01	0.07	0.11	0.12
5300.0	0.02	0.01	0.06	0.09	0.11

of Auger effect to double ionization resulting from vacancy in 2p shell (ionization threshold of 2p shell being 280 eV) have been considered in our calculations. As the fluorescence yield of the 2p shell of argon is very small of the order of 10^{-4} (see Krause [31]) and the value of the Auger yield is not available in literature we have assumed the Auger yield to be unity. We have shown 2p ionization cross-sections also in the figure and the table. As mentioned earlier, single ionization cross-sections for 2s shell are negligibly small and hence not considered in context of Auger effect.

First, we would like to discuss detailed comparison of direct double ionization cross-sections with the experimental data. At incident energies close to threshold the calculated results overestimate the cross-sections but they improve rapidly with increase in energy. It is seen that the cross-sections become within a factor of 2, 1.5 and 1.25 at incident energies 54.5 eV, 65 eV and 180 eV respectively. The improving trend continues and at 320 eV the theoretical and experimental cross-sections become almost equal. The peaks which appear at the same impact energy 108 eV in the region of direct double ionization in our calculation and experiment are of magnitudes $2.89 \times 10^{-16} \text{ cm}^2$ and 2.10×10^{-16} cm² respectively. It is remarkable that the position and magnitude of the predicted cross-section peak agree well with the corresponding experimental values. Thus we find that the calculated results show good agreement with the experimental data in the energy region 54.5–320 eV. This can be regarded as success of the present method for theoretical calculation of direct double ionization cross-sections. However, beyond 375 eV impact energy the calculated results are found to be smaller as compared to experimental values with increase in energy. At incident energies 1000 eV, 2000 eV and 3000 eV the calculated direct double ionization cross-sections are found to be less than two-third, half and two-fifth of the experimental cross-sections respectively. Further, the results at 4600 eV and 5300 eV become nearly one-third and one-fourth of the experimental values. Here we would like to mention that results calculated in the BEA become more and more accurate at increasing incident energies. Therefore the above mentioned trend of direct double ionization cross-sections strongly supports the idea of the contribution of Auger effect to double ionization crosssections.

Now we will examine the double ionization crosssections including contributions from Auger effect. It is seen that beyond 500 eV the increasing theoretical results become closer and closer to experimental data with increase in energy. The theoretical and experimental crosssections are found to be almost identical in the energy region 750–1520 eV. Beyond 1520 eV the experimental cross-sections are found to be slightly larger than the theoretical values but they always remain within a factor of 1.25 throughout the energy region 1520–5300 eV. It is interesting to note that the contributions of Auger effect become more than those of direct double ionization in the energy region 2000–5300 eV. From a critical comparison of the results it is apparent that the calculated double ionization cross-sections including contributions from Auger effect show reasonably good agreement with the experimental observations throughout the energy range investigated.

4 Conclusions

On the basis of the present calculations it is observed that consideration of ionization of 3p and 3s shells explains overall satisfactory agreement of electron impact single ionization cross-sections of argon with the experimental data. It is also concluded that the present method gives reasonable values of direct double ionization crosssections. It has been found that the calculated results of direct double ionization cross-sections cannot explain the experimental observations in the energy region where indirect physical processes are effective. Inclusion of contributions of Auger effect due to vacancy in 2p shell brings the theoretical results of double ionization crosssections in reasonably good agreement with the experimental data. The identification of the above mentioned shell which has been substantiated theoretically is an interesting feature of the present calculations.

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