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Double Ionization of Silicon and Phosphorus by Electron-Impact

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Abstract

I have calculated double ionization cross sections of silicon and phosphorus by electron impact using double binary encounter model. Hartree-Fock momentum distribution has been used for both the ejected electrons during collision process. Contributions from inner shell in the double ionization have been included in the calculations. The results obtained have been found in reasonably good agreement with the experimental observations.

Keywords: Double ionization, Electron impact, Shake off process, One step process, Two step process

1. Introduction

Electron impact ionization cross sections are essential quantities for modeling of a variety of processes of current interest e.g. low-temperature plasmas which are important in semiconductor processing and in destroying volatile organic compounds. Low temperature plasmas also modify the mechanical properties of surfaces, in material processing and in nuclear fusion reactors. Absolute ionization cross sections are also needed for obtaining quantitative gas densities from mass spectrometry measurements[1]. Moreover, electron impact multiple ionization (mainly double) of atoms plays a vital role in the understanding of plasma behaviour in astrophysics and controlled thermonuclear physics. The total ionization cross section for various targets due to the impact of electrons is one of the essential set of data required in a wide area of applications ranging from radiation science to laser physics. Therefore, any contribution to fill the gap in the knowledge of these data becomes important[2]. From an academic point of view, the study of dynamics of electron-atom inelastic scattering leads to a better understanding of the physical structure of the target and how the energy and momentum are transferred between the colliding particles during the process[3].

Experimentally the process of multiple ionizations (mainly double) of atoms due to electron impact has been extensively studied in low, intermediate and high energy region. Theoretical studies of electron impact double ionization of atoms are important due to the fact that these studies give the contributions of various ionization mechanisms to double ionization process. Three mechanisms are involved in double ionization of an atomic target by electron impact; the shake-off (SO), the two-step 1 $(TS1)$ and the two-step 2 $(TS2)$. In SO mechanism, the incident electron ejects one target electron leading to ionization of the target which is not an Eigen state. During relaxation another electron is ejected and the target is doubly ionized. In $TS1$ mechanism the incident electron ejects one target electron. The ejected electron in turn knocks out another target electron of the same atom resulting in its double ionization. In $TS2$ mechanism the incident electron makes collisions with two different

electrons in the same atom successively and ionizes both of them leading to double ionization of the atom[4].

Quantal calculations of electron impact double ionization cross section of atoms are very complicated and are limited to low atomic number targets and the system having outer shell configuration ns^2. Theoretically, the double ionization of an atom by electron impact may be described in the framework of a first order or a second-order theory. In the first order model, the transition matrix element contains the potential for the interaction between the colliding electron as one of the target electron and the correlation between the target electrons is responsible for double ionization process. In the second order process, the incident electron interacts successively with two target electrons and ejects them one by one. Nath etal[5] have reported the calculations of electron impact double ionization of Li^+ in the framework of a first order theory in which the standard correlation has been taken into account by using correlated wave function for the target atom whereas partial account of dynamic correlation has been considered in the first channel. Electron impact double ionization of an atom results in three continuum electrons moving in the Coulomb field of an ion which is a quantal coulomb four-body problem. The time-dependent close coupling (TDCC) method has been used to calculate total cross section for electron impact double ionization of He [6,7] and H^- [8]. The results show good agreement with experiments. Later on non-perturbative TDCC and perturbative distorted wave methods were combined to calculate total cross section for electron impact double ionization of Mg [9]. The same group of workers also reported their calculations of electron impact double ionization of beryllium[10] using non-perturbative TDCC method along with a newly developed R-matrix double pseudo state (RMDPS) method and found encouraging results. They have also performed the calculation of B^+ [11] on the similar line and found encouraging agreement with experiments.

Since application of rigorous quantal methods for calculation of electron impact double ionization cross sections of atoms is limited, some empirical formulae based on fitting parameters have been proposed for fast calculation of these cross sections[12,13]. On the other hand, double binary encounter model suggested by Gryzinski[14] has been quite successful in providing electron impact double ionization cross sections of a number of atoms and ions[15]. In this paper, we aim to report for the first time, calculation of electron impact double ionization cross section of silicon and phosphorus. These elements are widely used in semiconductor industry and electron impact ionization cross sections of these atoms are required for semiconductor fabrications[16]. The results so obtained agree well with available experimental results thereby validating the calculation[17]. We expect this work will encourage other workers to take up further study of the problem.

2. Theoretical Methods

According to Gryzinski's[14] double binary encounter model the electron impact double ionization cross section is given by

$$Q^{ii} = Q_{ej}^{ii} + Q_{sc}^{ii} \tag{1}$$

where Q_{ej}^{ii} is the cross section for $TS1$ process and Q_{sc}^{ii} that for $TS2$ process. Because of indistinguishability of electron, values of Q_{ej}^{ii} and Q_{sc}^{ii} are equal at every energy, hence the electron impact double ionization cross section of an atom is given by[18]:

$$Q^{ii} = 2\,\frac{n_e\left(n_e - 1\right)}{4\,\pi \bar{r}^2} \int_{t=0}^{\infty} \int_{U_i + U_{ii}}^{E_q} \sigma_{\Delta E}$$

$$\times \int_{t=0}^{\infty} \int_{U_{ii}}^{\Delta E - U_i} \sigma_{\Delta E'} f'(t) \sqrt{U_{ii}}\, d(\Delta E') dt$$

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

In the above equation

$n_e =$ number of equivalent electrons from which ejection of electron takes place.

$E_q =$ incident energy

U_i, U_{ii} = energy required for ejection of the first and second electron respectively

ΔE = energy transferred from the incident electron to the target electron in the first interaction

$\bar{r} = \frac{R}{n_e^{1/3}}$, R being the radius of the shell of the target atom.

The quantity $\sigma_{\Delta E}$ is the cross section for energy transfer ΔE which is given by

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

in which s and t are two dimensionless quantities defined as $s^2 = \frac{v_1^2}{v_0^2}$ and $t^2 = \frac{v_2^2}{v_0^2}$, where v_1 and v_2 are the velocities of incident and target electrons respectively in atomic units and $v_0^2 = u$ is ionization potential of the target electron in Rydberg.

The quantity ϕ is given by

$$\phi = Cos\left(\sqrt{\frac{1}{\left(s^2 + u\right)u}} \ln s^2 \right) \tag{4}$$

In equation (2) symbols $f(t)$ and $f'(t)$ denote the Hartree-Fock momentum distribution for the first and the second ejected electrons respectively. In the present work cross sections for ejection of both the electrons have been integrated numerically over the energy transfer and Hartree-Fock momentum distribution for the electrons. The Hartree-Fock radial wave function given by Clementi and Roetti[19] are used for the construction of momentum distribution for electrons and shell radii for electrons have been taken equal to quantum mechanical values of points of maximum radial probability density as reported by Desclaux[20].

3. Results and Discussion

The electron impact double ionization cross sections of silicon Si and phosphorus P are calculated using the crossed beam method, described in section 2. The calculated results have been presented in tables 1 and 2 and figures 1 and 2 respectively along with the experimental observations of Freund etal[17]. In the experiments, cross sections were measured with crossed-beam method in which target atoms were prepared by neutralizing singly charged ions by charge transfer. To the best of our knowledge, no other theoretical calculation for these processes has been reported so far.

Here we would like to mention that in case of direct double ionization of an atom by electron impact after the ejection of the first electron, if the collision is slow, the target will get sufficient time for rearrangement and therefore for the ejection of the second electron the binding energy and momentum distribution function for the target ion can be used. However, for fast collision the target will not get sufficient time for rearrangement and so the values of binding energy and momentum distribution function of the neutral atom is proper choice.

Table 1: Double Ionization of Silicon by Electron Impact

Impact Energy (eV)	**Present calculated cross section ($10^{-17}cm^2$)**			**Experimental ionization cross section[17] ($10^{-17}cm^2$)**
	Contribution from 3p shell	**Including contributions from 3p and 3s shells**	**Total ionization cross section**	
30.0	0.68	0.78	0.78	1.10
32.0	0.92	1.04	1.04	1.70
34.0	1.13	1.80	1.80	2.20
36.0	1.29	2.67	2.67	2.60
38.0	1.41	3.52	3.52	2.80
40.0	1.51	4.27	4.27	3.20
45.0	1.65	5.70	5.70	3.20
50.0	1.69	6.57	6.57	3.30
55.0	1.68	7.03	7.03	3.30
60.0	1.64	7.23	7.23	3.50
65.0	1.58	7.25	7.25	3.50
70.0	1.51	7.17	7.17	3.40
75.0	1.44	7.01	7.01	3.30
80.0	1.37	6.80	6.80	3.10
85.0	1.30	6.57	6.57	2.90
90.0	1.24	6.34	6.34	3.10
95.0	1.18	6.09	6.09	2.80
100.0	1.12	5.85	5.85	2.70
105.0	1.06	5.61	5.61	2.60
110.0	1.01	5.38	5.38	2.70
115.0	0.96	5.16	5.16	2.60
120.0	0.92	4.95	4.95	2.50
125.0	0.88	4.75	4.75	2.50
130.0	0.84	4.56	4.56	2.50
135.0	0.80	4.38	4.38	2.30
140.0	0.77	4.21	4.21	2.40
145.0	0.74	4.04	4.06	2.40
150.0	0.71	3.89	3.91	2.20
155.0	0.68	3.74	3.77	2.40
160.0	0.65	3.60	3.64	2.30
165.0	0.63	3.47	3.51	2.20
170.0	0.60	3.35	3.39	2.10
175.0	0.58	3.23	3.28	2.20
180.0	0.56	3.12	3.17	2.20
185.0	0.54	3.01	3.07	2.00

190.0	0.52	2.91	2.97	2.10
195.0	0.51	2.82	2.88	2.00
200.0	0.49	2.73	2.79	2.00

Table 2: Double Ionization of Phosphorus by Electron Impact

Impact Energy (eV)	Present calculated cross section ($10^{-17}cm^2$)			Experimental ionization cross section[17] ($10^{-17}cm^2$)
	Contribution from 3p shell	Including contributions from 3p and 3s shells	Total ionization cross section	
32.0	0.17	0.17	0.17	0.30
34.0	0.59	0.59	0.59	0.50
36.0	1.09	1.09	1.09	0.80
38.0	1.57	1.57	1.57	1.10
40.0	2.00	2.02	2.02	1.50
45.0	2.83	3.09	3.09	2.10
50.0	3.35	3.87	3.87	2.30
55.0	3.63	4.37	4.37	2.40
60.0	3.77	4.64	4.64	2.50
65.0	3.80	4.77	4.77	2.70
70.0	3.76	4.79	4.79	2.60
75.0	3.69	4.75	4.75	2.50
80.0	3.59	4.66	4.66	2.40
85.0	3.48	4.55	4.55	2.40
90.0	3.36	4.42	4.42	2.40
95.0	3.24	4.28	4.28	2.30
100.0	3.11	4.14	4.14	2.30
105.0	2.99	3.99	3.99	2.30
110.0	2.88	3.85	3.85	2.20
115.0	2.77	3.71	3.71	2.10
120.0	2.66	3.57	3.57	2.00
125.0	2.56	3.44	3.44	2.00
130.0	2.46	3.32	3.32	2.00
135.0	2.37	3.20	3.20	2.00
140.0	2.28	3.09	3.09	1.90
145.0	2.19	2.98	2.98	1.90
150.0	2.12	2.88	2.88	1.90
155.0	2.04	2.78	2.78	1.80
160.0	1.97	2.68	2.68	1.80
165.0	1.90	2.59	2.59	1.80
170.0	1.84	2.51	2.51	1.70
175.0	1.78	2.43	2.43	1.70
180.0	1.72	2.35	2.36	1.80
185.0	1.66	2.28	2.30	1.70
190.0	1.61	2.21	2.23	1.70
195.0	1.56	2.14	2.17	1.60
200.0	1.52	2.08	2.11	1.50

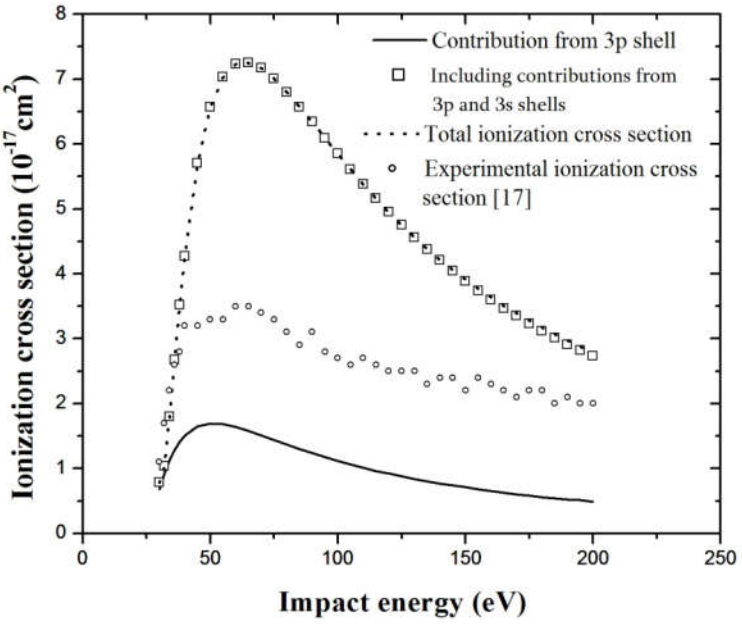

Fig. 1: Double ionization of Silicon by electron impact

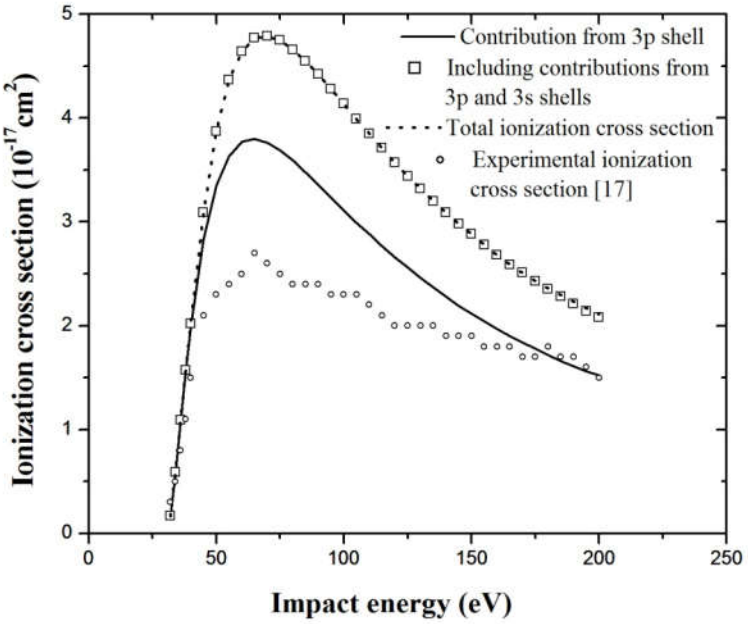

Fig. 2: Double ionization of Phosphorus by electron impact

In between these two limiting cases the rearrangement of the target after ejection of the first electron is partial. In case of heavy particle impact calculations, the concept of partial rearrangement of the target has been incorporated by Chatterjee and Roy[21] but unfortunately it is difficult to consider this in case of electron impact calculations[22].

In the present work we have assumed that after the ejection of the first electron from $3p$ shell the target attains the characteristics of the singly charged ion and so the ionization energy and the momentum distribution function for the singly charged ionic target have been used for the ejection of the second electron.

The valance shell of silicon is $3s^2 3p^2$. We have performed calculations in the energy range $30.0eV$ to $200.0eV$ for three cases (i) both the electrons are ejected from $3p$ shell $Q^{ii}(3p,3p)$ (ii) The first electron is ejected from $3p$ shell and the second from $3s$ shell $Q^{ii}(3p,3s)$ and (iii) the first electron is ejected from $3p$ shell and the second from $2p$ shell $Q^{ii}(3p,2p)$. Total double ionization cross section is taken as sum of the cross sections for these three processes.

$$Q^{ii} = Q^{ii}(3p,3p) + Q^{ii}(3p,3s) + Q^{ii}(3p,2p) \tag{5}$$

In case of calculation of $Q^{ii}(3p,3s)$ and $Q^{ii}(3p,2p)$ the factor $\frac{n_e(n_e-1)}{4\pi\bar{r}^2}$ appearing in equation (2) has been suitably modified. In these cases $n_e(n_e-1)$ has been replaced by $n_{e_1} \times n_{e_2}$ where n_{e_1} and n_{e_2} stand for the number of electrons in the shells under consideration. 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. The threshold energies for the processes corresponding to $Q^{ii}(3p,3p)$, $Q^{ii}(3p,3s)$ and $Q^{ii}(3p,2p)$ are $24.0eV$, $30.9eV$ and $137.46eV$ respectively[19]. The experimental cross sections shows a peak of magnitude close to $3.5\times10^{-17}cm^2$ at an impact energy between $60.0eV$ and $65.0eV$ which is double the threshold energy of $Q^{ii}(3p,3p)$. Our calculations show a peak of magnitude $1.69\times10^{-17}cm^2$ at $50.0eV$ when both the electrons are considered to be ejected from $3p$ shell. However, when the contributions from the process for ejection of the first electron from $3p$ shell and the second electron from $3s$ shell are included the position of the peak shifts to impact energy $65.0eV$ and the magnitude of peak becomes $7.25\times10^{-17}cm^2$. Contributions to the total double ionization cross section for the process $Q^{ii}(3p,2p)$ are negligible. Thus our calculations with contributions from $3p,3p$ process and $3p,3s$ processes reproduce the position of the peak almost correctly but the magnitude of the peak is about 2.07 times higher than the experimental peak. In the impact energy range $30.0eV$ to $34.0eV$ our cross sections are lower than the experimental values beyond which these become higher as compared to experimental cross sections. Between the impact energies, $55.0eV$ to $105.0eV$ the ratio of the present cross sections and the experimental cross sections is slightly more than 2.0 but otherwise the ratio is always less than 2.0 and agreement of the present results and experimental observations continues to improve with the increase in impact energy. The plot of experimental cross sections with energy shows number of small variations which might be due to the noise in the observed signal[17]. Moreover, the average slopes of experimental cross section curve and that of the present results appear to approach each other with increase in impact energy.

The results of phosphorus (P) in the energy range $32.0eV$ to $200.0eV$ are presented in table 2 and figure 2. In this case also we have performed three sets of calculations $Q^{ii}(3p,3p)$ with threshold energy $30.2eV$, $Q^{ii}(3p,3s)$ with threshold energy $38.78eV$ and $Q^{ii}(3p,2p)$ with threshold energy $168.48eV$ [19] while calculation of $Q^{ii}(3p,3s)$ and $Q^{ii}(3p,2s)$ the factor $\frac{n_e(n_e-1)}{4\pi\bar{r}^2}$ in equation (2) of section 2 has been modified as in case of silicon. The experimental cross sections show

a peak of magnitude $2.70\times10^{-17}cm^2$ at $65.0eV$ impact energy. The position of this peak is slightly higher than the threshold energy of the $Q^{ii}(3p,3p)$ process. Our $Q^{ii}(3p,3p)$ calculation show a peak of magnitude $3.80\times10^{-17}cm^2$ at $65.0eV$. The position of this peak coincides with that of the experimental peak but the magnitude of this peak is about 1.4 times higher. On including the contribution for $Q^{ii}(3p,3s)$ the position of the peak shifts to $70.0eV$ impact energy and its magnitude become $4.79\times10^{-17}cm^2$ which is about 1.77 times the magnitude of the experimental peak. As in case of silicon, the contribution for $Q^{ii}(3p,2p)$ is very small. In general, our calculated results are in reasonably good agreement with experiments throughout the energy range of investigations, the calculated results are always within a factor of 2.0 as compared to the experimental cross sections as reported earlier in literature also. However, both the experimental and theoretical values of ionization cross section results come closer and closer with the increase in impact energy both in magnitude and slope. In the case of silicon, the experiments show slight irregular variations in the cross section curve which might be due to noise in the received signal[17].

4. Conclusions

From the discussion given above it is concluded that the present method gives reasonably accurate values of electron impact direct double ionization of silicon and phosphorus. Here we would like to mention that in the previous similar calculation Jha etal[15] and Jha and Roy[18] have reported electron impact double ionization of argon and magnesium respectively. They have found that the calculated electron impact direct double ionization cross sections for both the systems are much smaller than the respective experimental results. However, on inclusion of contributions from single ionization cross section from inner shell followed by Auger effect leading to double ionization of the target the calculated cross sections show reasonably good agreement with the experimental results. This shows that increase of electron impact on argon and magnesium, indirect process has significant contribution in producing double ionization of the target. However, in this present case the calculated electron impact direct double ionization cross sections show reasonable agreement with experimental results indicating that in case of silicon and phosphorus due to electron impact double ionization is produced mainly due to direct double ionization and indirect process e.g., Shake Off process is expected to have insignificant contribution in producing double ionization of the atom. This is also supported theoretically as average value of Auger yield for $L-shell$ of silicon is approximately 0.025 and that of phosphorus is 0.029 [23].

Our method gives results closer and closer to experimental observations with the increase in impact energy. Further, only one set of experimental observation for electron impact double ionization cross sections for silicon and phosphorus are available. So the usefulness of the present method will be better ascertained if more experimental observations were available in the literature.

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