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Elastic cross-section calculations for electron collisions with XY_4 ($X = \text{Si, Ge}$; $Y = \text{H, F, Cl, Br, I}$) molecules

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Abstract

We report calculations of integral cross-section and differential cross-section for intermediate- and high-energy (20–2000 eV) elastic electron collisions with tetrahedral compounds of silicon and germanium: SiH_4 , SiF_4 , SiCl_4 , SiBr_4 , SiI_4 , GeH_4 , GeF_4 , GeCl_4 , GeBr_4 and GeI_4 . The calculations have been carried out using an independent atom model with static-polarization model potential. The present results are compared with available experimental and theoretical data. For collision energies higher than 100 eV, results of present theoretical calculations are in satisfactory agreement with experimental data.

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1. Introduction

The recent interest in electron-scattering cross-section data for tetrahedral compounds of silicon and germanium is partly related to the importance of these molecules in the chemistry of low-

temperature plasmas, manufacturing of semiconductors and other industrial processes. Moreover, due to their relatively simple geometry, the molecules of tetrahedral symmetry are convenient targets for testing different theoretical models applied to the electron-molecule scattering problem.

Experimental studies concerning elastic collision processes of electrons with XY_4 molecules are still rather very scarce. Elastic cross-section for electron collision with SiH_4 molecule was measured with crossed electron and molecular beam method by Tanaka et al. [1]. The same experimental group reported the differential cross-section (DCS) and elastic integral elastic cross-section

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(ICS) for electron scattering from GeH₄ molecule [2]. In both experiments energy of incident electrons did not exceed 100 eV.

Theoretical works on the electron scattering from silane (SiH₄) and germane (GeH₄) molecules were focused mainly on the low energy region [3–15] where ICS exhibit both Ramsauer minimum and a shape resonance maximum. For higher energy range elastic DCS and/or ICS for electron collisions with SiH₄ molecules were calculated using spherical-complex-optical-potential (SCOP) treated in variable phase approach (VPA) for electron energy between 30 and 400 eV by Jain [16] and from 10 to 5000 eV by Jain and Baluja [17]. Jain and Tripathi [18] employed relativistic approach to compute the DCS and ICS for e-SiH₄ collisions from 1 to 400 eV. Jain et al. [19] and Baluja et al. [20] using SCOP and VPA calculated elastic integral and DCS for GeH₄ molecules from 1 to 100 eV and 10 to 5000 eV, respectively. Kumar et al. [21] solved Dirac equation for e-GeH₄ scattering problem and obtained elastic cross-section for energies ranging from low up to 300 eV while Lee et al. [22] presented elastic integral and DCS for GeH₄ molecules calculated using the Schwinger iterative variational method in the fixed-nuclei, static exchange plus correlation–polarization approximation for energies between 0.2 and 100 eV.

Theoretical calculations of cross-sections for electron collisions with other tetrahedral compounds of silicon and germanium are less numerous than these for silane and germane molecules. Elastic scattering of low energy electrons from SiF₄ and SiCl₄ molecules was theoretically investigated by Tossell and Davenport [3] using multiple scattering X α calculations while Natalense et al. [23] and Varella et al. [24] reported elastic cross-sections for SiCl₄, SiBr₄ and SiI₄ molecules obtained in calculations in which Schwinger multichannel method (SMC) with norm-conserving pseudopotential was employed at energies below 40 eV. Recently, elastic cross-section for low energy (from 5 to 30 eV) electron collisions with GeCl₄ molecules has been calculated by Azevedo et al. [25] with the SMC method.

We are not aware any previous theoretical calculations related to electron collisions with GeF₄, GeBr₄ and GeI₄ molecules and any calculations of

electron elastic DCS and ICS for SiF₄, SiCl₄, SiI₄, SiBr₄, SiI₄ and GeCl₄ molecules for intermediate and high impact energies.

The aim of the present work was to calculate DCS and elastic ICS for electron scattering from tetrahedral compounds of silicon and germanium: SiH₄, SiF₄, SiCl₄, SiBr₄, SiI₄, GeH₄, GeF₄, GeCl₄, GeBr₄ and GeI₄, for impact energies ranging from tens of electronvolts up to 2000 eV. In the following section we present main assumptions of the used theoretical method and numerical procedures. In this presentation we adopted atomic units in which $e = m = \hbar = 1$, although all results of our calculation, presented and discussed in the next sections, are given in the SI units.

2. Theory

The calculations have been carried out using an independent atom method (IAM) [26–28] with static-polarization model potential. In this approximation the electron–molecule collision problem is reduced to electron–atom collision problem under the following assumptions [27]: (i) each atom of the molecule scatters independently, (ii) any redistribution of atomic electrons due to the molecular binding is unimportant so that each atom scatters as if it were free, (iii) multiple scattering within the molecules is negligible. Above assumptions seem to be valid for collisions with fast electrons only. This approach was successfully employed for calculations of elastic, momentum transfer as well as total cross-sections for intermediate- and high-energy electron scattering from many diatomic and polyatomic molecular targets [29–46].

In the IAM method the DCS for elastic electron scattering on molecule, taking into account all possible orientations of the intermolecular axis in the space, is given as

$$\frac{d\sigma}{d\Omega} = \sum_i^N \sum_j^N f_i(\theta, k) f_j^*(\theta, k) \frac{\sin(sr_{ij})}{sr_{ij}}, \quad (1)$$

where N is the number of atoms within molecule, θ is the scattering angle and $f_i(\theta, k)$ and $f_j(\theta, k)$ are complex scattering amplitudes due to the i th and j th atom of the molecule, respectively. $s = 2k \sin$

$(\theta/2)$ is the magnitude of the momentum transfer during the collision and k is the wave number of the incident electron. The distance between the i th and j th atom is denoted as r_{ij} . From Eq. (1) it follows that the DCS for elastic electron scattering from molecules of tetrahedral symmetry can be derived as

$$\begin{aligned} \left(\frac{d\sigma}{d\Omega}\right)_{XY_4} &= \left(\frac{d\sigma}{d\Omega}\right)_X + 4\left(\frac{d\sigma}{d\Omega}\right)_Y \left(1 + 3\frac{\sin(sr_{YY})}{sr_{YY}}\right) \\ &+ 8[\operatorname{Re}f_X(\theta, k)\operatorname{Re}f_Y(\theta, k) \\ &+ \operatorname{Im}f_X(\theta, k)\operatorname{Im}f_Y(\theta, k)]\frac{\sin(sr_{XY})}{sr_{XY}}, \end{aligned} \quad (2)$$

where $(d\sigma/d\Omega)_X$ and $(d\sigma/d\Omega)_Y$ are DCS for electron scattering from X and Y atoms, respectively, while $\operatorname{Re}f_X(\theta, k)$, $\operatorname{Re}f_Y(\theta, k)$, $\operatorname{Im}f_X(\theta, k)$ and $\operatorname{Im}f_Y(\theta, k)$ are real and imaginary parts of the scattering amplitude on the X and Y atoms, respectively. It follows from the optical theorem that the elastic ICS for electron scattering on the molecule in this approximation is given by

$$\begin{aligned} \sigma(E) &= \frac{4\pi}{k}\operatorname{Im}f(s=0, k) \\ &= \frac{4\pi}{k}\sum_{i=1}^N\operatorname{Im}f_i(\theta=0, k) \\ &= \sum_{i=1}^N\sigma_i(E), \end{aligned} \quad (3)$$

where $\sigma_i(E)$ is the ICS of the i th atom of the molecule, and $E = k^2/2$ is the energy of the incident electron.

To obtain the elastic electron–atom cross-sections and atomic scattering amplitudes we employed partial wave analysis and solved numerically the radial Schrödinger equation

$$\left[\frac{d^2}{dr^2} - \frac{l(l+1)}{r^2} + k^2 - 2(V_{\text{stat}}(r) + V_{\text{polar}}(r))\right]u_l(r) = 0, \quad (4)$$

under the boundary conditions

$$u_l(0) = 0, \quad u_l(r) \xrightarrow{r \rightarrow \infty} A_l \hat{j}_l(kr) - B_l \hat{n}_l(kr), \quad (5)$$

where $\hat{j}_l(kr)$ and $\hat{n}_l(kr)$ are the spherical Bessel–Riccati and Neumann–Riccati functions, respectively. $V_{\text{stat}}(r)$ is the static potential of the atom

determined following the procedure of Salvat et al. [47],

$$V_{\text{stat}}(r) = -\frac{Z}{r} \sum_{i=1}^3 a_i \exp(-\beta_i r), \quad (6)$$

where Z is the nuclear charge and β_i and a_i are the parameters, determined by an analytical fitting procedure to Dirac–Hartree–Fock–Slater self-consistent data [47]. The polarization potential $V_{\text{polar}}(r)$ was expressed in the form proposed by Padial et al. [48],

$$V_{\text{polar}}(r) = \begin{cases} v(r) & r \leq r_c \\ -\alpha/2r^4 & r > r_c \end{cases}, \quad (7)$$

where $v(r)$ is the free-electron-gas correlation energy [49] and α is the static electric dipole polarizability of atom. The r_c is the first crossing point of the $v(r)$ and $-\alpha/2r^4$ curves [50]. The values of r_c are listed in Table 1. The phaseshifts δ_l are connected with the asymptotic form of the wave function, $u_l(r)$, by

$$\tan \delta_l = \frac{B_l}{A_l}. \quad (8)$$

The scattering amplitudes for electron scattering on the atom were obtained using the following equation:

$$\begin{aligned} f(\theta, k) &= \frac{1}{2ik} \sum_{l=0}^{\infty} (2l+1)(e^{2i\delta_l} - 1)P_l(\cos \theta) \\ &\approx \frac{1}{2ik} \sum_{l=0}^{l_{\text{max}}} (2l+1)(e^{2i\delta_l} - 1)P_l(\cos \theta) \\ &+ f^{(B)}(\theta, k), \end{aligned} \quad (9)$$

where $P_l(\cos \theta)$ are Legendre polynomials and $f^{(B)}(\theta, k)$ is the Born scattering amplitude which for potential of the form (7) may be expressed as

Table 1
The crossing point r_c

Atom	r_c (Å)
H	1.57
F	1.42
Si	2.79
Cl	2.14
Ge	2.93
Br	2.42
I	2.87

$$\begin{aligned}
 f^{(B)}(\theta, k) &= \pi\alpha k \sum_{l=l_{\max}+1}^{\infty} \frac{P_l(\cos\theta)}{(2l-1)(2l+3)} \\
 &= \pi\alpha k \left(\frac{1}{3} - \frac{1}{2} \sin \frac{\theta}{2} \right. \\
 &\quad \left. - \sum_{l=1}^{l_{\max}} \frac{P_l(\cos\theta)}{(2l-1)(2l+3)} \right), \quad (10)
 \end{aligned}$$

in the presented calculations $l_{\max} = 100$.

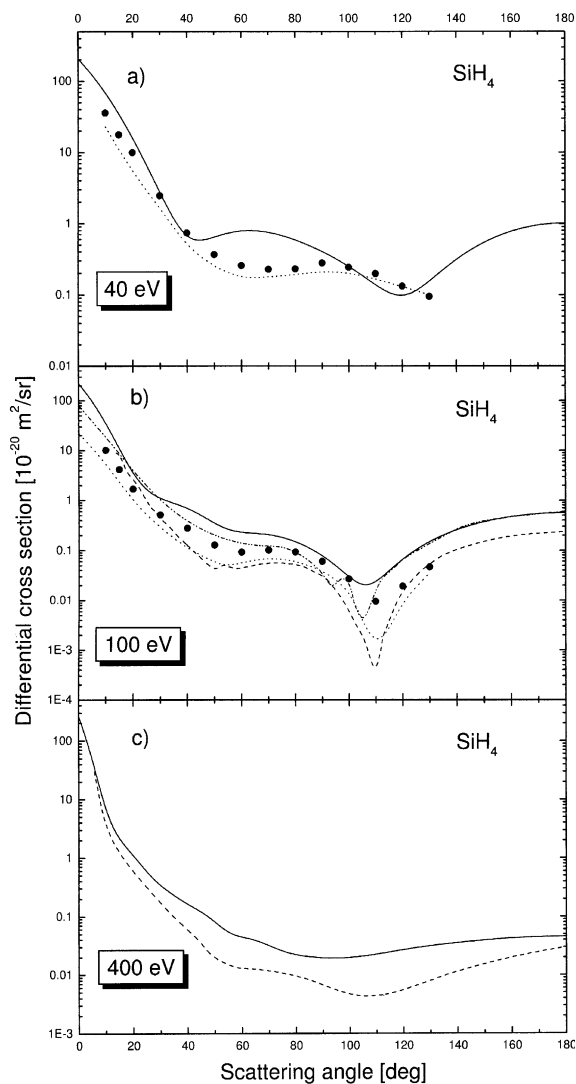


Fig. 1. DCS for e^- - SiH_4 collisions. (a) 40 eV. Experiment: (●) [1]. Theory: (—) present results; (···) [1]. (b) 100 eV. Experiment: (●) [1]. Theory: (—) present results; (···) [1]; (---) [16] (---) [18]. (c) 400 eV. Theory: (—) present results; (---) [16].

Finally, the scattering amplitude can be expressed as

$$\begin{aligned}
 f(\theta, k) &= \frac{1}{2ik} \sum_{l=0}^{l_{\max}} (2l+1)(e^{2i\delta_l} - 1)P_l(\cos\theta) \\
 &\quad + \pi\alpha k \left(\frac{1}{3} - \frac{1}{2} \sin \frac{\theta}{2} - \sum_{l=1}^{l_{\max}} \frac{P_l(\cos\theta)}{(2l-1)(2l+3)} \right). \quad (11)
 \end{aligned}$$

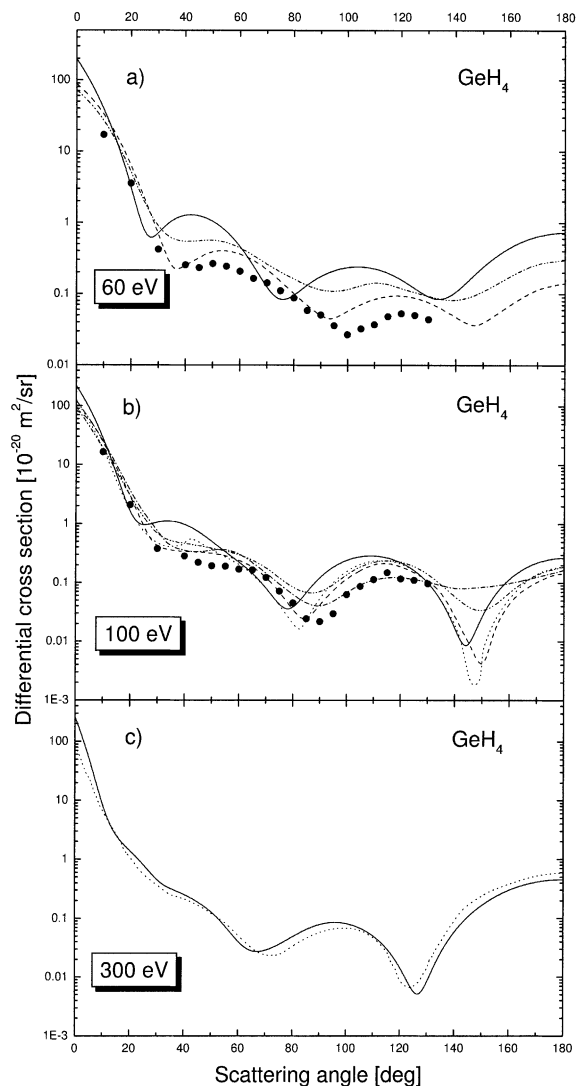


Fig. 2. DCS for e^- - GeH_4 collisions. (a) 60 eV. Experiment: (●) [2]. Theory: (—) present results; (---) [19]; (---) [22]. (b) 100 eV. Experiment: (●) [2]. Theory: (—) present results; (---) [19]; (---) [2]; (···) [21]; (---) [22]. (c) 300 eV. Theory: (—) present results; (···) [21].

DCS for elastic electron scattering from atoms were calculated according to

$$\frac{d\sigma}{d\Omega} = |f(\theta, k)|^2, \tag{12}$$

while ICS for elastic electron-atom scattering were derived from the expression

$$\sigma = \frac{4\pi}{k^2} \left(\sum_{l=0}^{l_{\max}} (2l + 1) \sin^2 \delta_l + \sum_{l=l_{\max}+1}^{\infty} (2l + 1) \sin^2 \delta_l^{(B)} \right). \tag{13}$$

3. Results

3.1. Differential cross-section

DCSs for e^- -SiH₄ collisions at energy 40, 100 and 400 eV calculated in the reported work are presented and compared with other available theoretical and experimental data in the Fig. 1(a)–(c),

respectively. Generally, the shape of all cross-sections presented in Fig. 1 is similar. However, for $E = 40$ eV observed structures of our cross-section are shifted towards lower angles (of about 20 degrees) in comparison with experimental data. For higher impact energies (100 eV and more) much better agreement of present results with experimental data of Tanaka et al. [1] and results of other calculations [1,16,18] can be observed. Presented results have nearly the same angular distribution as the other data. On the other hand, our calculations give the cross-sections which exceed experimental data and other theoretical cross-sections.

Results of our DCS calculations for electron scattering from germane (GeH₄) molecules are presented in Fig. 2(a) for energy of 60 eV, (b) for 100 eV, and (c) for 300 eV. It can be easily seen that for 60 eV the present cross-section has the same shape as previously obtained experimental and theoretical results although all structures in the cross-section function are shifted towards lower scattering angles. For higher energies, agreement

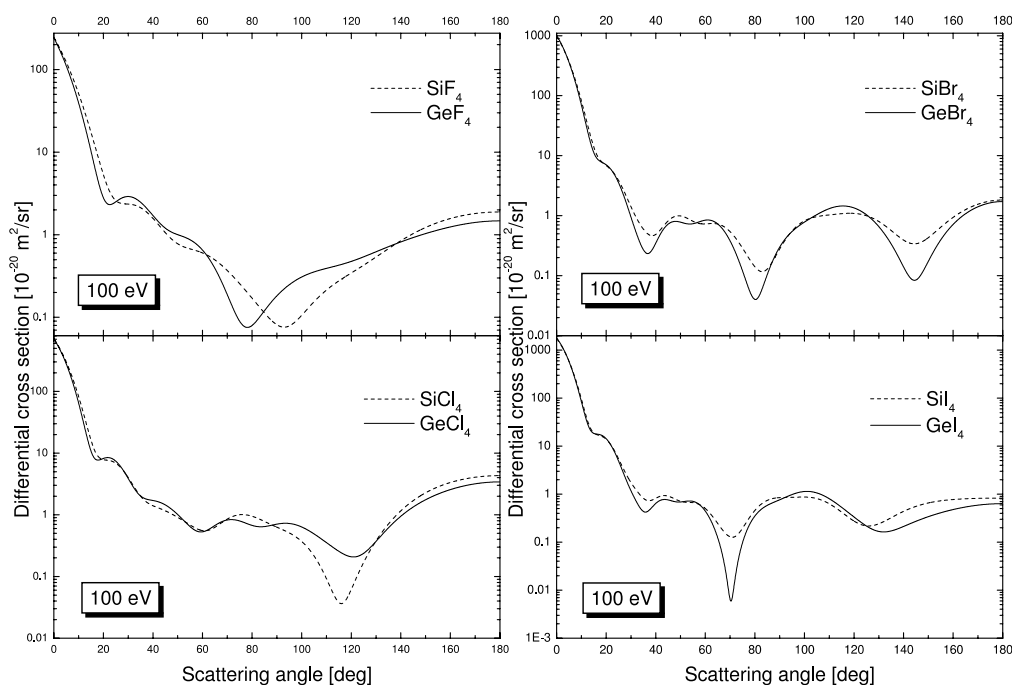


Fig. 3. DCS for electron collisions with SiF₄, SiCl₄, SiBr₄, SiI₄, GeF₄, GeCl₄, GeBr₄ and GeI₄ molecules for 100 eV.

with the other data, with respect to the shape and positions of the local minima and maxima, is much better, especially with relativistic calculations of Kumar et al. [21] at 300 eV.

DCSs for electron scattering from XF_4 , XCl_4 , XBr_4 and XI_4 ($X = \text{Si, Ge}$) molecules for collision energy of 100 eV are presented in Fig. 3. No other data, both experimental and theoretical, are available for comparison. It can be easily seen that DCSs for these molecules have more reach structures in angular dependence than those for SiH_4 and GeH_4 . Moreover, it seems that cross-section shape depends on the kind of outermost atoms of

the molecules rather than on the central one. Similar behaviour was observed in experimental total cross-section for molecules of tetrahedral symmetry [51].

3.2. Integral cross-section

Elastic ICS for electron collisions with XY_4 molecules are listed in Table 2. We found via comparison with experimental total cross-sections [51–54], that for SiF_4 , SiCl_4 , GeF_4 and GeCl_4 molecules the cross-sections calculated in the present work have reasonable magnitude for energies

Table 2

Elastic ICS for electron collisions with tetrahedral XY_4 ($X = \text{Si, Ge}$; $Y = \text{H, F, Cl, Br, I}$) molecules in units of 10^{-20} m^2

Energy (eV)	ICS									
	SiH_4	SiF_4	SiCl_4	SiBr_4	SiI_4	GeH_4	GeF_4	GeCl_4	GeBr_4	GeI_4
20	24.29					23.07				
25	20.65					16.14				
30	18.13					12.89				
40	14.83					10.07				
50	12.75	24.36	40.56	24.64	44.96	8.85	20.47	36.66	20.75	41.06
60	11.31	21.26	35.75	24.05	42.95	8.17	18.11	32.61	20.91	39.80
70	10.25	19.08	32.25	23.93	39.43	7.71	16.54	29.71	21.39	36.89
80	9.43	17.46	29.64	23.90	36.30	7.36	15.39	27.57	21.83	34.24
90	8.76	16.19	27.63	23.81	33.97	7.08	14.51	25.95	22.13	32.29
100	8.21	15.16	26.03	23.64	32.34	6.83	13.79	24.66	22.27	30.96
110	7.74	14.31	24.72	23.40	31.18	6.61	13.19	23.59	22.27	30.05
120	7.33	13.59	23.61	23.09	30.32	6.41	12.67	22.69	22.18	29.40
140	6.66	12.41	21.81	22.39	29.09	6.05	11.81	21.21	21.78	28.49
160	6.12	11.48	20.39	21.64	28.17	5.74	11.10	20.01	21.26	27.78
180	5.68	10.72	19.23	20.91	27.35	5.45	10.50	19.01	20.69	27.13
200	5.30	10.09	18.24	20.21	26.59	5.20	9.99	18.14	20.11	26.49
220	4.98	9.54	17.38	19.56	25.86	4.97	9.53	17.37	19.55	25.85
250	4.58	8.85	16.28	18.67	24.81	4.67	8.93	16.37	18.76	24.90
300	4.05	7.93	14.80	17.37	23.21	4.24	8.12	14.99	17.57	23.40
350	3.64	7.22	13.62	16.27	21.80	3.89	7.47	13.87	16.53	22.05
400	3.32	6.64	12.65	15.33	20.57	3.61	6.93	12.94	15.62	20.85
450	3.06	6.17	11.84	14.51	19.49	3.37	6.48	12.15	14.82	19.80
500	2.84	5.76	11.14	13.79	18.54	3.16	6.09	11.46	14.12	18.87
600	2.49	5.11	10.00	12.59	16.96	2.83	5.46	10.34	12.94	17.31
700	2.22	4.61	9.10	11.63	15.70	2.58	4.96	9.46	11.98	16.06
800	2.02	4.21	8.38	10.84	14.67	2.38	4.56	8.73	11.19	15.03
900	1.85	3.87	7.77	10.17	13.81	2.21	4.23	8.13	10.53	14.17
1000	1.71	3.59	7.27	9.60	13.08	2.07	3.96	7.63	9.96	13.44
1100	1.60	3.36	6.83	9.11	12.44	1.96	3.72	7.19	9.47	12.80
1200	1.50	3.15	6.45	8.68	11.89	1.86	3.51	6.81	9.04	12.25
1400	1.34	2.82	5.83	7.96	10.97	1.69	3.17	6.18	8.31	11.32
1600	1.22	2.55	5.33	7.38	10.22	1.57	2.90	5.68	7.73	10.57
1800	1.12	2.34	4.93	6.91	9.61	1.46	2.69	5.28	7.25	9.95
2000	1.05	2.17	4.60	6.51	9.10	1.38	2.51	4.94	6.85	9.43

higher than 50 eV. Below this energy the ICS increases rapidly with decreasing of collision energy and at low energies (~ 10 eV) exceeds the total cross-section. One may therefore expect that similar behaviour appears also for SiBr_4 , SiI_4 , GeBr_4 and GeI_4 molecules for which experimental data are not yet available. However, for SiH_4 and GeH_4 molecules we found that the computed ICS are in good agreement with experimental measurements of Tanaka et al. [1] and Dillon et al. [2] even at 20 eV.

In Fig. 4 we compared our calculated elastic ICS with ICS obtained from experiment [1] and that computed by Jain [16]. For comparison we plotted also total cross-section calculated by Jain et al. [18] and total cross-sections measured by Szymtkowski et al. [52] and Zecca et al. [55]. Our calculations are in satisfactory agreement with experimental data of Tanaka et al. [1] for lower energies and lie close to theoretical cross-section of Jain [16] for higher energies. However, at 100 eV, the discrepancy between theoretical cross-sections and experimental data of Tanaka et al. [1], presented in Fig. 4 amounts over 50%. It can be partially attributed to the procedure of extrapolation of experimental DCS towards low ($<10^\circ$) and high ($>130^\circ$) angles used in [1] to obtain ICS. It is worth noting that ICS presented by Tanaka et al. is higher in its maximum than total cross-section [52] at the same energy.

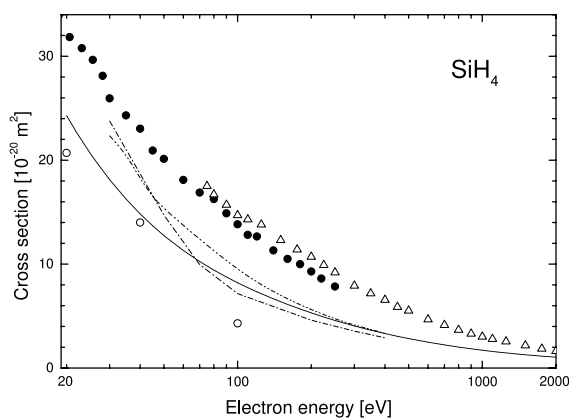


Fig. 4. Cross-sections for e^- - SiH_4 collisions. Calculated: (—) integral elastic, present results; (---) elastic ICS [16]; (- · - ·) total cross-section [18]. Experimental: (○) elastic cross-section [1]; (●) absolute TCS, [52]; (△) absolute TCS, [55].

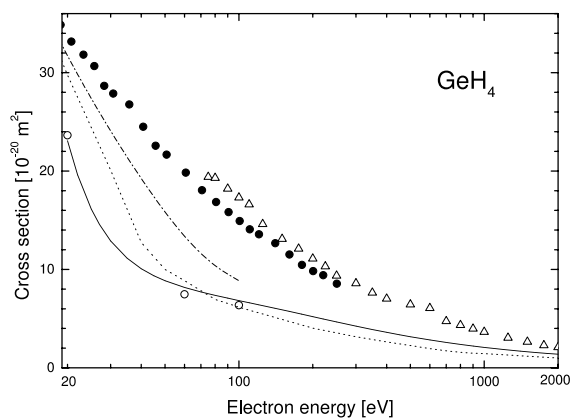


Fig. 5. Cross-sections for e^- - GeH_4 collisions. Calculated elastic ICS: (—) present results; (···) [20]; (- · - ·) [22]. Experimental: (○) elastic cross-section, [2]; (●) absolute TCS, [56]; (△) absolute TCS, [57].

In Fig. 5 we compare elastic ICS for electron collisions with GeH_4 molecules obtained in the present calculations with experimental ICS data of Dillon et al. [2] and calculations of Baluja et al. [20] and Lee et al. [22]. We plotted also absolute total cross-sections measured by Mozejko et al. [56] and by Karwasz [57]. Our calculations are in very good agreement with measurements of Dillon et al. [2] and for energies higher than 100 eV also with calculation of Baluja et al. [20]. Results of Lee et al. [22] are higher than our results and that of Baluja et al. [20] as well as experimental data of Dillon et al. [2].

From our calculations we estimated that elastic processes for the XY_4 ($X = \text{Si, Ge}$; $Y = \text{H, F, Cl}$) molecules constitute up to 60% of the absolute total cross-sections for intermediate energies (100–250 eV) and no more than 65% for the highest examined energy.

4. Summary

We calculated with simply method, namely IAM elastic ICS and elastic DCSs for electron collisions with tetrahedral XY_4 ($X = \text{Si, Ge}$; $Y = \text{H, F, Cl, Br, I}$) for a wide energy range (from tens of eV up to 2000 eV). Presented results are above 100 eV in good agreement with available experimental cross-section data and with results of

more sophisticated calculations for electron scattering from tetrahedral compounds of silicon and germanium.

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