Low- and intermediate-energy total electron scattering cross sections for SiH$_4$ and GeCl$_4$ molecules

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Abstract. Absolute total cross sections for electron collision with SiH$_4$ and GeCl$_4$ molecules have been measured in a linear attenuation experiment for impact energies ranging from 0.6 to 250 eV.

Good agreement of the present e$^-$–SiH$_4$ results with other experimental data has been found in respect of the energy dependence of the total cross section (TCS). Discrepancies in magnitude are only apparent for the lowest and the highest impact energies applied. Comparison is also made with theoretical calculations.

The electron-scattering TCS for GeCl$_4$ shows two distinct resonant maxima: one at 1.7 eV, occurring over a narrow energy range, the other—much broader—near 9 eV. No TCS data for GeCl$_4$ are available for comparison.

1. Introduction

The literature on electron scattering from molecules includes a considerable number of experimental works on the SiH$_4$ molecule. The earlier reports of Steele et al (1962), Saalfeld and Svec (1963), Hess and Lampe (1966), Potzinger and Lampe (1969) and the more recent work of Chatham et al (1984) have dealt with the electron-impact ionization and dissociation of the monosilane molecule. The formation of negative ions in electron–SiH$_4$ collisions has been reported by Ebinghaus et al (1964) and Giordan (1983). Among e$^-$–SiH$_4$ experiments those employing the swarm technique, in which electron transport parameters in the monosilane gas have been determined, are especially numerous (for extensive references see Morgan 1992). A momentum transfer cross section derived from the swarm data reveals a deep Ramsauer–Townsend (RT) minimum between 0.3 and 0.4 eV, followed by a broad maximum centred near 2–3 eV (Ohmori et al 1986, Mathieson et al 1987, Kurachi and Nakamura 1989, Wang et al 1991, Nagpal and Garscadden 1994).

Experiments on vibrational excitation of silane (Mathieson et al 1987, Tronc et al 1989, Tanaka et al 1990) demonstrated that the sharp increase of the cross section above the RT minimum is related to the formation of a shape resonance state through accommodation of the incident electron in an orbital of the target molecule. Dissociative excitation of silane by electron impact has been reported by Perrin et al (1982), Perrin and Aarts (1983) and by Tsurubuchi et al (1992).


Investigations on the electron–GeCl₄ scattering processes are less numerous and can be briefly summarized as follows. Electron dissociative capture has been studied experimentally by Pabst et al (1977) and, recently, by Guillot et al (1996). The latter have also carried out experiments on electron transmission spectroscopy (in the 0–7 eV electron-energy range) and on inner-shell electron-energy-loss spectroscopy. The emission spectra resulting from electron impact have been recorded by Lambert et al (1988). It should be noted, that intensities of the hitherto examined processes accompanying the e⁻–GeCl₄ scattering have not been given in the absolute scale.

In this paper, we present measurements of absolute grand TCS for electrons scattered by SiH₄ and GeCl₄ molecules at energies ranging from 0.6 to 250 eV.

2. Experimental

To determine electron scattering TCS we have used the transmission method, which relates cross section at a given impact energy, $E$, to the transparency of the target at a given pressure, $p$, for a beam of projectiles (cf Bederson and Kieffer 1971).

An electron beam was produced with an electron gun, and energy selected by passing through a 127° electrostatic cylindrical deflector. A quasimonoenergetic electron beam left the monochromator with an energy spread of 70 meV and intensity between 1 and 100 pA. A system of electron lenses aligned electrons and directed them into a scattering chamber. The portion of the electron beam which left the reaction volume through the exit orifice was energetically discriminated with a retarding field analyser and, finally, detected by the Faraday cup. The magnetic field in the region of electron optics was reduced to 0.1 µT.

In a perfect transmission experiment—i.e. when (i) the projectile beam is monoenergetic, parallel and very narrow, (ii) the concentration of target molecules, $n$, is low enough for interactions with projectiles to be single only, and (iii) only those projectiles are registered which have not interacted with target molecules, while (iv) the solid angle subtended by the detector is very near zero—the TCS, $Q(E)$, is related to attenuation of the electron beam as follows:

$$Q(E) = \left( \int_a^b n(x) \, dx \right)^{-1} \ln \frac{I_o(E)}{I_s(E)},$$

where $I_o(E)$ and $I_s(E)$ are intensities of detected transmitted electron currents of energy $E$ in the presence or absence of the target in the scattering volume, respectively; $n(x)$ is the target density at point $x$ of the electron pathway, while integration is performed over the entire path of the electron beam where $n(x) \neq 0$.

The conceptually simple problem of determining a TCS through measurements of a few physical quantities becomes a difficult task when one wishes to obtain the TCS with great accuracy.
The inevitable effusion of target particles across orifices, through which electrons enter and leave the reaction chamber, means that a number of noticeable scattering events may also occur outside the scattering cell, notwithstanding the fact that the applied geometry of the electron optics ensures good pumping of the background volume near the orifices; consequently, determination of the value of the integral in expression (1)

$$\int_a^b n(x) \, dx$$

is a critical point in the evaluation of TCS. Following the effective scattering pathlength calculations of Nelson and Colgate (1973), we have found that in the present experiment the integral (2) may be, within an accuracy of 0.4%, replaced by the product \( nL \), where \( L = 30.5 \) mm is the geometrical distance between orifices of the scattering chamber and \( n \) is the target density number in the centre of the scattering cell.

The density number, \( n \), is usually determined from its relation to target pressure in the scattering volume and target temperature. The absolute gas pressure in the scattering cell was measured with a capacitance gauge (MKS) calibrated by the manufacturer before the present experiment. Since the temperature of the capacitance manometer head in our experiment, \( T_m = 322 \) K, differs by a few degrees from the temperature \( T_{sc} \) of the scattering cell, a correction of pressure readings due to the thermal transpiration effect has been necessary. The gas density, \( n \), was therefore determined from the corrected (Knudsen 1910) ideal gas formula:

$$n = \frac{p_m}{k \sqrt{T_m T_{sc}}}$$

where \( p_m \) is pressure according to a baratron and \( k \) is the Boltzmann constant. Following Baldwin and Gaerttner (1973) we have found that in the present experiment the \( n \) values obtained from the simple formula (3) differ by no more than 0.5% from values obtained from more sophisticated empirical expressions (e.g. Poulter et al 1983).

The pressure of the sample in the scattering volume was kept below 100 mPa at low-impact energies and did not exceed 200 mPa at the highest energies applied, where cross sections are distinctly lower. Those upper limits on pressure in the collision cell proved sufficient to ensure negligibility of multiple-scattering events.

In ‘target in’ operational mode, the pressure in the region of electron optics (monitored with an ionization gauge) was lower than that in the scattering cell by nearly three orders of magnitude. However, changes in target pressure in the reaction chamber by a few orders of magnitude lead to variations in the density of target molecules occurring also in the region of electron optics and may alter its transmission properties, thus influencing measured cross section. Variations of partial target pressure in the background may also strongly affect emission from the filament, as has been noticed in the case of silane. In order to lessen these effects, the target sample was supplied alternately into a collision cell or in the background in such a way that conditions along the path of the electron beam outside the scattering cell were stable throughout the experiment, irrespective of whether the target was present or absent in the collision chamber.

Another essential source of erroneous results in all transmission experiments, which systematically lowers the measured cross sections, is imperfect discrimination with respect to particles which have undergone scattering in small-angle forward directions, due to finite angular resolution of the detector system.

The difference, \( \Delta Q(E) \), between the actual cross section, \( Q_e(E) \), and the measured
cross section, $Q_\text{m}(E)$, due to forward scattering is given by the following expression:

$$
\Delta Q(E) = \left( \int_a^b n(x) \, dx \right)^{-1} \int_a^b n(x) \, dx \int_0^{\Delta\Omega_1} \left( \frac{dQ(E)}{d\Omega} \right) \, d\Omega,
$$

(4)

where $\Delta\Omega_1$ is the solid angle at which the detector’s surface is visible from point $x$ of the beam path, where scattering occurs, and $dQ(E)/d\Omega$ is a differential cross section for electron scattering (elastic + inelastic). The difference $\Delta Q(E)$ increases with impact energy.

In order to estimate the effect of small-angle scattering on the measured TCS, angular distribution functions of scattered electrons at energies under study are needed. Unfortunately, pertinent data are mostly available for elastic scattering only, and are limited to only a few impact energies. A reasonable, though rather overvalued, estimation of the corrective term (4) would be the product of the elastic differential scattering cross section at zero angle and the solid angle subtended by the electron detector as seen from the centre of the scattering cell: $(dQ_{el}/d\Omega)_0\Delta\Omega_1$. Using the differential SiH$_4$–electron scattering cross sections measured by Tanaka et al. (1990), extrapolated to zero angle, we conclude that for the present scattering geometry ($\Delta\Omega_1 \sim 1 \text{ msr}$) $\Delta Q$ should not exceed 1% even at the highest applied impact energy, 250 eV. A similar value of $\Delta Q$ might be expected for the GeCl$_4$ molecule. The data presented in the present work have not been corrected to allow for this effect.

To reduce the number of inelastically scattered electrons, which might be accepted by the collector together with transmitted unscattered electrons, a retarding field element was applied before the Faraday cup collector. This enabled us to discriminate electrons with kinetic energies that differed from the impact energy by no more than tenths of an eV.

The experimental energy scale of the electron beam was determined by the voltage applied to the source, and the absolute energies were established with an accuracy of $\pm 50 \text{ meV}$ by reference to the resonant oscillatory structure of N$_2$ around 2.3 eV.

The absolute measurements of the temperature of the target, $T$, its pressure, $p$, and electron beam intensities, $I$, were performed with high-accuracy devices.

Control of the measurement cycle, data accumulation and data processing were performed entirely by a computer.

At each energy, individual cross section measurements were made over a wide range of electron optical and gas pressure conditions. The results proved to be independent (within the limits of statistical uncertainties) of the pressure of the target, the background pressure and the intensity of the electron beam. The final value of the TCS at each particular impact energy is an average of a large number of data obtained in an independent series (6–24) of individual runs (usually 10 in a series).

Statistical uncertainties (one standard deviation of weighted mean values) for the e$^-$–SiH$_4$ scattering TCS are approximately near 1.5% below 1.5 eV and do not exceed 1% at higher energies, whereas for GeCl$_4$ they are well below 1% over the entire energy range studied. The direct sum of potential individual systematic errors has been estimated to be up to 9% below 2.5 eV for SiH$_4$ and below 5 eV for GeCl$_4$, decreasing below 3% between 10 and 100 eV, and increasing again up to 4% at the highest applied energies. The relatively large uncertainties at the lowest impact energies for both molecules examined are mainly due to sharp changes of the cross section functions in this energy range, where a small drift in energy may bring about a relatively great change in cross section.

The SiH$_4$ sample (of Matheson purity) was used directly from its cylinder. The GeCl$_4$ (Fluka, at least 99.5% pure) vapours, were purified before use by subjecting them to numerous freeze–pump–thaw cycles at the liquid N$_2$ temperature to remove air and other impurities not frozen at this temperature.
Table 1. Absolute TCS measured for electron impact on SiH₄ and GeCl₄ molecules in units of $10^{-20}$ m².

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3. Results

The resulting absolute TCS for SiH₄ and GeCl₄ molecules in the electron impact energy range of 0.6–250 eV are summarized in table 1.

3.1. Monosilane, SiH₄

Figure 1 compares the present absolute experimental data for the SiH₄ molecule with the absolute measurements of Wan et al (1989) in the low-energy range (up to 12 eV) and with the absolute results of Zecca et al (1992) (between 75 and 250 eV); normalized data of Sueoka et al (1994) are also included. All the presented TCS are in good agreement with regard to shape, while there are certain discrepancies in magnitude. The present results are higher than those of Wan et al (1989) at all energies of overlap (below 2 eV by more than 15%), but have remained within the limits of the combined uncertainties of both experiments.
At the same time, above 100 eV, the results of Zecca et al (1992) are above ours and this discrepancy increases steadily with energy up to about 15% at 250 eV. Such disagreement is rather disturbing, as—so far—other results obtained at the laboratories in Gdańsk and in Trento for this energy range have lain within the limits of declared uncertainties. It seems unlikely that the present results should be so much lower due to the finite angular resolution of the detector. Observed disagreements may partly result from the use of a magnetic field in TCS experiments of Wan et al (1989), Zecca et al (1992) and Sueoka et al (1994) which may bring about additional systematical experimental errors, often difficult to estimate (Golden 1978).

It may also be interesting to note that the sum of the elastic cross section (Tanaka et al 1990) and the ionization TCS (Perrin et al 1982, Chatham et al 1984) at 100 eV constitutes almost 70% of the grand TCS, in satisfactory agreement with the majority of results for other molecules. At the same time, the sum of the elastic cross section at 100 eV and the dissociation TCS at the same energy (Perrin and Aarts 1983) yields a value in excess of the grand TCS, which suggests an erroneous estimation of the dissociative attachment cross section.

In figure 2, the present experimental e\(^-\)–SiH\(_4\) results are shown together with some of the available theoretical cross section calculations. Computations of Jain and Thompson (1991), making use of an exact-exchange and parameter-free model of polarization potential, reproduce quite well the general shape of the experimental curve with respect to energy. However, the calculations yield the resonance structure that is narrower and noticeably larger at the maximum. This discrepancy could be lessened if vibrations of the nuclei were included in the calculations (cf Lynch et al 1979). Theoretical position of the maximum is clearly shifted by about 1 eV towards the higher energies, in comparison with the experiment. Calculations of others (Yuan 1988, Winstead and McKoy 1990) reproduce the behaviour of the cross section around 3 eV quite adequately, though certain discrepancies with the experiment are apparent.

Jain and Baluja (1992) and, more recently, Jiang et al (1995) have calculated TCS
Electron–SiH\textsubscript{4} and electron–GeCl\textsubscript{4} scattering

Figure 2. Computed cross sections for e\textsuperscript{−}–SiH\textsubscript{4} scattering: ---, Yuan (1988); ......, Winstead and McKoy (1990); -- --, Jain and Thompson (1991); ---, Jain and Baluja (1992); ·······, Jiang \textit{et al} (1995), compared with experiment: ⋄, grand total, present results.

The model complex optical potential. Results of Jain and Baluja above 20 eV are in very good agreement with the present experimental data. Below 20 eV, however, when energy decreases, the calculated TCS increases more than in the experiment and the discrepancy reaches about 20% at 10 eV. The calculations of Jiang \textit{et al} (1995), which ignore molecular geometry (by using the additivity rule to obtain molecular cross sections from atomic cross sections), differ considerably from the experiment throughout the comparable energy region; the disagreement is the highest at 10 eV (70%) and decreases with energy increasing to 30% around 250 eV.

3.2. Tetrachlorogermane, GeCl\textsubscript{4}

Absolute electron scattering TCS for GeCl\textsubscript{4} versus impact energy is displayed in figure 3. The general energy dependence of the measured TCS is dominated by two distinct maxima and is similar to that observed for some other tetrachlorides (for CCl\textsubscript{4} see Holst and Holtsmark 1931, Jones \textit{et al} 1986, Wan \textit{et al} 1989, Szmytkowski \textit{et al} 1992, for SiCl\textsubscript{4} see Wan \textit{et al} 1989).

At the lowest energies applied in this experiment, a steep increase of cross section is visible from 36 \times 10\textsuperscript{−20} m\textsuperscript{2} at 0.6 eV up to the first maximum value of 57 \times 10\textsuperscript{−20} m\textsuperscript{2}, centred near 1.7 eV, followed by a rapid decrease of cross section to the minimum of 47 \times 10\textsuperscript{−20} m\textsuperscript{2} at 3 eV. This sharp, narrow feature might be attributed to the existence, close to 1.7 eV, of a short-lived resonant state created by the capture of an extra electron into the unoccupied orbital of the molecule.

The position of the maximum TCS on the energy scale coincides with the resonant effects noticed by Pabst \textit{et al} (1977) near 1.9 eV in the dissociative attachment experiment and by Guillot \textit{et al} (1996), at 1.72 eV, in the electron current transmitted through the GeCl\textsubscript{4} vapour.

Past the minimum at about 3 eV, we observe another sharp increase of the cross section up to its second maximum value of 80 \times 10\textsuperscript{−20} m\textsuperscript{2} near 10 eV. On the left side of this hump, a
shoulder is clearly visible near 6 eV. Numerous processes are energetically accessible in the region of this very broad feature, which makes detailed analysis very difficult. However, calculations carried out for other molecules (Lynch et al 1979) indicate that such broad maximum around 10 eV could be partly associated with the existence of a number of weak overlapping resonances. Experimental support in favour of such assumption for the e−–GeCl4 scattering comes from studies of dissociative electron attachment to this molecule (Pabst et al 1977, Guillot et al 1996). Guillot et al (1996) have also recorded some relatively narrow resonant structures around 5.6 eV in the electron transmission spectra. They associated observed features with core-excited resonance created when the extra electron is temporarily captured to the parent neutral excited state or, alternatively, with shape resonance, when an electron is quasibound to a molecule in its electronic ground state.

![Figure 3](image1.png)

**Figure 3.** Absolute electron–GeCl4 scattering grand TCS measured in this work. Error bars in selected points represent the total (systematic plus statistical) experimental uncertainties.

![Figure 4](image2.png)

**Figure 4.** Energy dependence of the TCS for selected XY4 molecules: • GeCl4 present; ◆ CCl4 Szmytkowski et al (1992); ■ GeH4 Mozejko et al (1996); ▼ SiH4 present; ▲ CH4 Zecca et al (1991). Straight lines across the points are the regression lines.
For impact energies above the second maximum, TCS falls monotonically with increasing energy and reaches a value of $25 \times 10^{-20}$ m$^2$ at the highest applied energy of 250 eV.

Above 40 eV, the dependence of the $e^-–$GeCl$_4$ TCS on energy can be nicely approximated with the $\sigma \sim E^{-0.5}$ function, which means that the TCS at intermediate energies is just proportional to the time needed by the electron to cover the distance in the vicinity of a target molecule. A similar energy dependence for the 50–250 eV range has been exhibited by TCS for some other XY$_4$ targets (see figure 4) measured in our laboratory (Zecca et al. 1991, Szmytkowski et al. 1992, Mozejko et al. 1996).

To our knowledge, no experimental electron scattering total cross sections or theoretical calculations for GeCl$_4$ have been available for comparison, so far.

As the hitherto published experiments have not explicitly explained the origin of the observed resonant-like structures, further and more detailed studies on the $e^-–$GeCl$_4$ scattering are necessary. It may also be interesting to extend cross section measurements towards lower energies, where one would expect Ramsauer minimum behaviour.

Acknowledgments

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