LETTER TO THE EDITOR

Electron collisions with germane (GeH_4) molecules. Absolute total cross section measurements from 0.75 to 250 eV

Paweł Możejko, Grzegorz Kasperski and Czesław Szmytkowski Faculty of Applied Physics and Mathematics, Technical University of Gdańsk, 80-952 Gdańsk, Poland

Received 1 April 1996

Abstract. Absolute total cross section for electron collisions with GeH₄ molecules has been measured in a linear electron transmission experiment for impact energies between 0.75 eV and 250 eV. Experimental cross section function versus energy reveals a resonant-like broad enhancement with maximum between 3 and 4 eV, monotonically decreasing at higher energies.

The recent interest in electron-scattering cross section data for the germane molecule is partly related to the importance of this compound in the chemistry of low-temperature plasmas and the physics of semiconductors. Accurate experimental results are also urgently needed to verify the reliability of theoretical models used for computations.

The relative simplicity in the theoretical treatment of the e^- -GeH₄ scattering problem, due to nearly spherical symmetry of the molecule, encourages the testing of different models of electron–molecule interaction. Most of the calculated cross sections are elastic integral and/or momentum transfer cross sections (Winstead *et al* 1991, Jain *et al* 1991, Dillon *et al* 1993, Bettega *et al* 1993, 1995 and Kumar *et al* 1995). Total (elastic + absorption) cross sections in the electron energy range of 10–5000 eV have been calculated by Baluja *et al* (1992) using the spherical–complex–optical potential (SCOP) model.

Experimental investigations of processes induced by electron collisions with germane molecules have been relatively rare so far. More effort in this field has been concentrated on dissociative effects (Perrin and Aarts 1983, Perrin and Schmitt 1984 and Lloret *et al* 1991). Moreover, Maracci *et al* (1989) obtained electron-impact excited M-shell Auger spectra of the germane molecule, while Dillon *et al* (1993) determined integral elastic cross sections from their measurements of angular distributions at energies from 1 to 100 eV. Most recently, Karwasz (1995) has measured the first absolute electron scattering total cross sections at intermediate (75–4000 eV) impact energies. We are not aware of any low energy experimental e⁻–GeH₄ total cross section data.

The objective of the present study is to provide experimental total cross sections for the GeH₄ molecule at impact energies ranging from low to intermediate.

The experiment reported here was carried out with an electron spectrometer working in the linear transmission configuration. The apparatus and procedures employed in this work have been described previously by Krzysztofowicz and Szmytkowski (1995) and will be only briefly mentioned here.

An electron beam was formed by a 127° cylindrical electrostatic dispersing element and a system of electron lenses. Energy-selected electrons entered a collision chamber filled with target molecules. The transmitted electrons were energetically discriminated with a retarding field element and finally detected by a Faraday cup.

The method employed is based on measurements of electron-beam attenuation through the target gas (cf Bederson and Kieffer 1971). The total cross section, Q(E), at a given energy E was derived by measuring intensities of the transmitted electron current in the presence (I_p) , and absence (I_o) of the target molecules in the scattering cell and by applying the Bouguer–de Beer–Lambert formula:

$$Q(E) = \frac{1}{nL} \ln \frac{I_o(E)}{I_p(E)};\tag{1}$$

the other quantities being length of the scattering cell (L = 30.5 mm) and absolute number density n of the target, determined from absolute measurements of the gas-target pressure (< 0.2 Pa) and temperature, taking into account the thermal transpiration effect (Knudsen 1910).

Table 1.	Absolute tota	1 cross section	s (TCS)	measured	for	electron	impact	of GeH ₄	molecule
in units o	of 10^{-20} m ² .						-		

Energy (eV)	TCS	Energy (eV)	TCS	Energy (eV)	TCS
0.75	12.5	4.3	56.2	25	30.7
0.85	12.8	4.8	54.4	28	28.6
1.0	17.3	5.3	53.0	30	27.9
1.1	20.2	5.8	51.1	35	26.8
1.2	23.3	6.3	50.6	40	24.5
1.3	26.2	6.8	49.8	45	22.6
1.4	31.6	7.3	49.4	50	21.7
1.5	34.0	7.8	48.6	60	19.8
1.6	35.8	8.5	48.2	70	18.0
1.7	37.2	9.0	47.2	80	16.8
1.8	38.1	9.5	46.7	90	15.8
1.9	40.5	10	46.5	100	14.9
2.0	42.3	11	45.2	110	14.1
2.2	44.3	12	43.4	120	13.6
2.4	47.5	13	42.2	140	12.7
2.6	51.2	15	39.9	160	11.5
2.8	53.2	17	37.5	180	10.5
3.1	55.4	19	34.8	200	9.83
3.5	57.9	21	33.2	220	9.41
3.8	58.8	23	31.8	250	8.55

The electron-energy scale for the total cross section function was calibrated by reference to the oscillatory resonant structure in the transmission current around 2.3 eV in molecular nitrogen.

Commercially supplied sample GeH_4 gas (99.99% from Air Products) was used without further purification.

Final values of the total cross section are the weighted means of average cross sections obtained in independent series (6–24) of individual runs (6–10 in a series) for the same impact energy. Statistical uncertainties reach about 1.5% below 2 eV and do not exceed 0.5% at higher energies. The direct sum of potential individual systematic errors has been

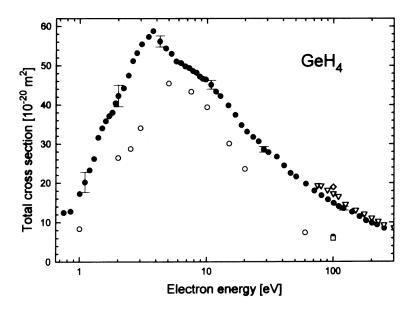


Figure 1. Experimental electron–GeH₄ scattering cross sections: Grand total: (\bullet), present absolute (the error bars correspond to overall experimental uncertainties estimated at selected points); (∇), absolute experimental of Karwasz (1995). Total: (\square), for ionization, and (\Diamond), for dissociative attachment, Perrin and Aarts (1983). Elastic: (\circ), Dillon *et al* (1993).

estimated to be up to 7% on the steep slope of the cross section function below 1.5 eV, 2–3% between 2 and 100 eV, and 4% elsewhere.

The total cross section determined in the present experiment is shown in figure 1 together with other available experimental data. Figure 2 compares the computed total and integral elastic cross sections with the presently measured total cross sections. Table 1 presents our total cross section results in numerical form.

The most striking feature of the measured e^- -GeH₄ total cross section function, clearly illustrated in figure 1, is its dramatic increase from 12×10^{-20} m² at 0.8 eV up to nearly 59×10^{-20} m² at the 3.8 eV maximum. This pronounced structure is partly attributable to the existence, between 3 and 4 eV, of a short-lived resonant state created by a capture of an extra electron into the lowest unoccupied orbital of the molecule. Evidence of resonant effects between 2 and 3 eV has been noticed by Dillon *et al* (1993) in their elastic and vibrationally inelastic experiments. A shape-resonant feature around 5 eV has also been reported in the elastic calculations of Jain *et al* (1991) and explained in terms of d-wave electron scattering with the formation of a temporary negative-ion state. The shoulder in the cross section, apparent on the high-energy side of the very broad maximum, between 6 and 11 eV, is probably caused by the presence of very broad overlapping resonances in this energy range.

For electron energies greater than 10 eV, the measured total cross section becomes systematically smaller with increasing impact energy; at 250 eV it decreases to 8.6×10^{-20} m². The elastic integral cross section measured by Dillon *et al* (1993) and the total electronionization cross section obtained in the plasma experiment of Perrin and Aarts (1983) are of nearly the same magnitude at 100 eV, and their sum constitutes almost 80% of the grand total cross section at this impact energy, in resonable accord with results obtained

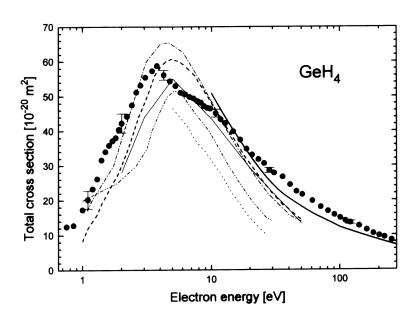


Figure 2. Computed cross sections for e⁻-GeH₄ scattering. Total: (—), (elastic + absorption), Baluja *et al* (1992); Elastic: (·····), Schwinger multichannel (SMC) approach, Winstead *et al* (1991); (- · - -), spherical effective potential, Jain *et al* (1991); (— · —), SMC, Bettega *et al* (1993, 1995); (——), continuum multiple scattering, Dillon *et al* (1993); (— · —), relativistic, Kumar *et al* (1995); Experimental: (•), grand total, present.

for other molecules. However, the sum of the total dissociation cross section at 100 eV (Perrin and Aarts 1983) and the elastic cross section (Dillon *et al* 1993), exceeds the grand total cross section by more than the combined experimental uncertainties, which suggests that the value of the dissociative attachment cross section, especially in channels leading to neutral fragments, has been overestimated.

As can be seen in figure 1, our results are in good accord with the recent intermediate-energy total cross section measurements of Karwasz (1995), in the common energy range of both experiments. A discrepancy of up to 15% between these total cross sections present between 75 and 110 eV is partly attributable to beam instability effects in this energy range observed in the Trento apparatus (Zecca *et al* 1991). It should be noted that March *et al* (1994) have been quite successful in their phenomenological prediction of the maximum total cross section value $(53 \times 10^{-20} \text{ m}^2)$; the divergence from the present experiment is no more than 10%.

The overall shape of all the cross section functions, both experimental (figure 1) and theoretical (figure 2), is similar throughout the overlapping energy range. However, there are some differences in the position of the maximum: the theoretical elastic cross section's maximum is shifted by about 1–1.5 eV towards higher energies as compared with our experimental total cross section. In general, the location of the maximum depends on the theoretical approach used for solution of the e⁻–GeH₄ scattering problem. It is also noteworthy that some calculations, especially relativistic ones (Kumar *et al* 1995), have considerably overestimated elastic cross sections around the maximum; some of them are

more than the present grand total cross sections.

The only e⁻-GeH₄ total (elastic + absorption) cross section calculation of Baluja *et al* (1992) reasonably corresponds with the present experiment; at 10 eV, their theoretical cross sections exceed our data by about 10% while being constantly less then the present results above 30 eV, by slightly more than 15%.

GeH₄ molecule is isovalent with CH₄ and SiH₄ molecules, for which a Ramsauer minimum has been observed close to 0.4 eV (Ferch *et al* 1985, Lohmann and Buckman 1986) and 0.3 eV (Wan *et al* 1989), respectively. A similar minimum may also be expected in the e⁻-GeH₄ cross section. A reference to an elastic cross section minimum at 0.1 eV can be found in the theoretical work of Jain *et al* (1991). March *et al* (1994), while analysing the shape of the total cross section function for 'quasi-spherical' molecules, predicted a Ramsauer minimum in the e⁻-GeH₄ cross section at 0.185 eV. Unfortunately, this energy region could not be included in the reported experiment.

We would like to thank Professor A Zecca for some facilities. This work was partially sponsored by Komitet Badań Naukowych.

References

Baluja K L, Jain A, Di Martino V and Gianturco F A 1992 Europhys. Lett. 17 139-44

Bederson B and Kieffer L J 1971 Rev. Mod. Phys. 43 601-40

Bettega M H F, Ferreira L G and Lima M A P 1993 Phys. Rev. A 47 1111-8

Bettega M H F, Natalense A P P, Lima M A P and Ferreira L G 1995 J. Chem. Phys. 103 10566-70

Dillon M A, Boesten L, Tanaka H, Kimura M and Sato H 1993 J. Phys. B: At. Mol. Opt. Phys. 26 3147-58

Ferch J, Granitza B and Raith W 1985 J. Phys. B: At. Mol. Phys. 18 L445-50

Jain A, Baluja K L, Di Martino V and Gianturco F A 1991 Chem. Phys. Lett. 183 34-9

Karwasz G P 1995 J. Phys. B: At. Mol. Opt. Phys. 28 1301-9

Knudsen M 1910 Ann. Phys., Lpz. 31 205-29

Krzysztofowicz A M and Szmytkowski Cz 1995 J. Phys. B: At. Mol. Opt. Phys. 28 1593-602

Kumar P, Jain A K and Tripathi A N 1995 J. Phys. B: At. Mol. Opt. Phys. 28 L387-92

Lloret A, Oria M, Séoudi B and Abouaf-Marguin L 1991 Chem. Phys. Lett. 179 329-33

Lohmann B and Buckman S J 1986 J. Phys. B: At. Mol. Phys. 19 2565-70

Maracci F, Platania R, De Sueoza A C and De Sueoza G G B 1989 Chem. Phys. 133 291-5

March N H, Zecca A and Karwasz G P 1994 Z. Phys. D 32 93-100

Perrin J and Aarts J F M 1983 Chem. Phys. 80 351-65

Perrin J and Schmitt J P M 1984 Chem. Phys. Lett. 112 69-74

Wan H-X, Moore J H and Tossell J A 1989 J. Chem. Phys. 91 7340-7

Winstead C, Hipes P G, Lima M A P and McKoy V 1991 J. Chem. Phys. 94 5455-61

Zecca A, Karwasz G, Brusa R S and Grisenti R 1991 J. Phys. B: At. Mol. Opt. Phys. 24 2737-46