Electron interactions with tetrahydrofuran

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Abstract. In this paper, we summarize our recent experimental and theoretical results on electron scattering from gaseous tetrahydrofuran (THF). Electron-impact ionization and total scattering cross sections were determined experimentally for energies between 50-5000 eV. Electron energy loss spectra were measured in the keV range using a transmission beam technique and for smaller energies (15-50 eV) with a crossed-beam apparatus. Using an optical potential method assuming the screening-corrected additivity rule, total, elastic and inelastic cross sections including dipole interactions were calculated (1eV - 10keV) in order to complement the experimental data. Elastic differential cross sections were also obtained. An empirical approximation to the inelastic angular distributions based on differential cross sections is proposed. The available integral and differential cross sections and energy loss distributions in the range 1 eV - 10 keV are combined into a table of recommended electron interaction cross sections with THF.

1. Introduction
It is well known [1] that high energy radiation produces abundant secondary electrons (~4·10⁴ per MeV of energy deposited), which are the main source of the energy transfer map and radiation damage in biological tissues. Even electrons with sub-ionisation energies can produce damage, in terms of DNA strand breaks and molecular dissociation, more efficiently than the traditionally considered mechanism of direct ionisation of the medium (Sanche and colleagues [2]). Unfortunately, DNA is not itself readily amenable for the studies needed to determine a complete set of electron interaction data, for example when it comes to modelling radiation effects in a simulation. As a consequence, moieties of DNA like tetrahydrofuran (THF), as well as water, can serve as model systems for attempting to build up the requisite data bases for track simulations in matter [3].

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Here we summarize recent measurements and calculations by our group on different aspects of electron scattering from THF and present new energy loss distributions and elastic differential cross sections. In section 2, we compare the experimental and theoretical total cross sections (CS). Section 3 deals with ionization cross sections. Elastic differential cross sections are presented in section 4. Finally, in section 5 electron energy loss (EEL) spectra for two different energy ranges are presented. Based on these distributions, we derive electronic excitation cross sections and propose an empirical approximation for inelastic angular distributions in the low energy range. In the discussion and conclusion section, we aim to establish a set of recommended electron-THF cross section data and present these in resulting tables.

2. Total scattering cross sections

Absolute total electron scattering cross sections for gaseous THF were obtained previously in the range 50eV – 5keV, by measuring the electron beam attenuation in a transmission beam experiment [3]. In brief, the apparatus employed consisted of an electron gun, an interaction chamber containing the target gas, and a hemispherical energy analyzer followed by an electron channel multiplier as a detector. Three electrostatic quadrupoles along the beam's path were used for focussing and aligning the electrons. The experimental uncertainty of the present measurement is estimated to be below 5%, according to the analysis of the main error sources carried out in [4].

The optical potential method described in previous papers [5,6] has been used to calculate differential and integral elastic, as well as integral inelastic, electron-THF scattering CS for energies up to 10 keV. This calculation was based on an independent atom model (IAM) but using a screening-corrected additivity rule (SCAR) and included recent adjustments [6,7] especially important in the low energy region. Processes involving nuclear motion were neglected in this calculation. The present method considered inelastic scattering as being due to electron-electron interaction processes; only those arising from electronic excitation were taken into account, so that vibrational excitations are ignored. However, rotational excitation was included by taking into account dipole scattering contributions.

![Figure 1. Total scattering cross sections, obtained experimentally in Madrid, compared to the results of our IAM-SCAR calculations. Experimental total cross sections published by Zecca et al. [9] and Mozejko et al. [8] et al. are also included.](image-url)
The total scattering cross sections obtained both experimentally and theoretically [3] are plotted in figure 1. An excellent agreement between our experiment and calculation can be observed in the keV range. For lower energies, differences were found to be ≤ 10%. On the other hand, the present experimental results are within 7% of those presented by Mozejko et al. [8] for the overlapping energy range. For low energies (< 15 eV), we find a large difference between our calculation and experiments by other groups. This is not surprising since an independent atom model, even with screening corrections, cannot be expected to give accurate predictions for such low energies. On the other hand, the large dipole moment of the THF molecule enhances rotational excitation particularly for low energies (see figure 1). This is reflected in a higher theoretical total CS due to the fact that theory deals with an ideal system with all target molecules in their ground state. This is, however, not a realistic premise for an experimental system at room temperature where rotationally excited levels are populated.

3. Ionization

Electron-impact ionization was determined previously [3] for energies between 50 eV and 5 keV, by combining simultaneous electron and ion measurements with a time-of-flight analysis of the ionic fragments. In brief, electrons were generated by an emitting filament and then driven into a gas cell by a pulsed voltage. This collision chamber featured two perpendicular apertures, each leading to a drift tube. Positive ions and secondary electrons were simultaneously detected in the two opposite directions, by two-stage micro-channel plate assemblies. The beam current transmitted through the gas cell was also measured in order to normalize the ionization signal and thus obtain ionization cross sections. The total electron-impact ionization cross sections for THF [3] can be seen in figure 1. The associated uncertainty is estimated to be about 7% including uncertainties in the normalization procedure. We observe very good agreement of about 6% with the theoretical (binary-encounter-Bethe) data of [10].

4. Elastic differential cross sections

![Figure 2](image)

**Figure 2.** Comparison of our IAM-SCAR elastic DCS (black solid line) to results obtained in other experiments [12-15] and calculations [16].

In addition to integral scattering cross sections, elastic differential cross sections (DCS) were also calculated with the IAM-SCAR method. A comparison of our results with different experiments and the calculations of Winstead and McKoy [16] is displayed in figure 2. Similar to what has been observed for other molecules such as methane [11] and ethylene, these theoretical angular distributions agree well with the experimental data of Milosavljevic et al. [12] in the higher energy range. At 50 eV (their upper limit), we also find good agreement with the results of Colyer et al. [13] both in shape and magnitude. Below about 30 eV, increasing discrepancies become apparent between the experimental [13-15] and theoretical data. Our calculation does not predict the steep increase of the elastic cross
section at forward scattering angles (below 30°-40°) which is present in the experimental results. Agreement does become better, however, when including dipole interactions in our calculation. A further difference lies in the localization of the global minimum of the angular distributions, which appears shifted towards higher angles in the theoretical DCS at energies < 30 eV.

5. Electron energy loss distributions, electronic excitation and inelastic DCSs

EEL distributions were measured for two incident electron energy ranges. From 300 eV up to 1000 eV, spectra were recorded in Madrid in the same apparatus described in section 2 and used for the total CS measurements. In this case for the energy loss measurements, the electrostatic quadrupoles were employed to tilt the beam's trajectory and thus obtain a mixture of small-angle scattered electrons at the analyzer entrance while avoiding the primary beam. In figure 3, the average distributions obtained for different energies are presented. As can be seen, variations between those spectra are minimal and do not follow any clear trend with energy. The average over all distributions (right panel of figure 3) yields a mean energy loss for the processes covered (electronic excitation, ionization, and neutral dissociation) of 26.7 eV. Note the excellent agreement, taking into account the poorer resolution, in the overlapping range (onset of the inelastic region) of this average distribution with the average of the 50 eV spectra recorded at Flinders University.

![Figure 3](image1.png)

**Figure 3.** Energy loss spectra obtained for the incident electron energy range 300-1000 eV. The right plot shows the average of all distributions measured. The dashed line illustrates how it could be adapted for use with a lower incident electron energy (30 eV).

![Figure 4](image2.png)

**Figure 4.** Example of the energy loss spectra obtained at Flinders University at 20 eV (left panel) and 50 eV (right panel). Three energy bands I, II and III of Rydberg...
states, as denoted in the figure, were analyzed separately.

For the incident energies 15 - 50 eV and energy losses up to 11 eV, spectra were recorded at Flinders University for well-defined angles from 15° to 90° [12]. The energy resolution achieved in those experiments was 50-60 meV. Figure 4 shows some spectra obtained at 20 eV and 50 eV. In order to derive differential cross sections for electronic excitation, the energy loss region was first divided in three bands of Rydberg states as denoted in the plot. A fitting procedure adapted to accommodate the particular spectroscopy of THF [14], was then used to yield the ratio of the DCS for the band of interest (band I, band II, or band III), to that for the elastic DCS [13]. In this way, differential electronic excitation cross sections were derived [12]. Integral electronic excitation cross sections were determined using a molecular phase shift analysis technique for extrapolation to 0° and 180°.

Since experimental inelastic differential cross sections are in many cases only available for a restricted range of scattering angles and incident energies, applications like electron transport simulations often have to rely on approximations in spite of the growing efforts to obtain accurate inelastic angular distributions. This is why the present differential energy loss spectra were further examined, in search of an empirical formula approximating inelastic scattering behaviour based on elastic DCS (generally more easily available).

When inspecting the experimental angular distributions of electrons scattered inelastically (after having excited the target molecule electronically), we observed that as a general trend, differences in magnitude (maxima and minima) become smaller for increasing energy loss $\Delta E$ and scattering into large angles becomes more important. This means that that the steep increase in the DCS at forward angles present for low energy losses, similar to the elastic DCS, is much less prominent for larger energy losses relative to the incident energy. An electron having excited a higher energy level or caused ionization, is thus more likely scattered by a larger angle than an electron having excited the lowest state. A diagram of this qualitative trend is displayed in figure 5.

**Figure 5.** Schematic of the general behaviour observed for elastic and inelastic angular distributions as a function of the energy loss.

A suitable approximation should reproduce these findings. For reasons of mathematical consistency, we also require our distribution to tend towards the elastic DCS for $\Delta E \rightarrow 0$. Taking those points into account, we propose an approximation of the inelastic angular distributions by:

\[
\frac{\partial^2 \sigma}{\partial \Omega \partial \Delta E} \propto \left(\frac{\partial \sigma}{\partial \Omega}\right)^{\Delta E/E}_{el},
\]

for a given incident energy $E$, where $\sigma$ is the cross section and $\Omega$ a solid angle. Absolute values (DCS) can be obtained by normalization to the corresponding integral CS. One can easily see that this approximation yields the elastic CS ($\partial \sigma/\partial \Omega)_e$ for $\Delta E=0$ and produces an isotropic distribution for the extreme case of complete energy loss, $\Delta E=E$. In fact it thus represents a gradual transition between two popular approximations, namely the supposition of isotropic scattering and the direct use of elastic DCS for inelastic events.

This formula is of course a very rough approximation to a complex problem encompassing several, physically diverse, collision processes. However, it is obviously more accurate than the two before-mentioned methods, while being easy to implement in any simulation and not requiring extensive
calculations. The distributions resulting for 15 eV, 30 eV and 50 eV as well as the corresponding experimental points are displayed in figure 6. The main difference to the method considering momentum transfer, used previously for the cases of water and methane [20,11], is the relatively larger weight for the backscattering. When comparing the two methods, we noted an improvement in emulating the experimental points with the present approximation. A quantitative evaluation shows a mean deviation from the experimental data of 17% (present formula) vs. 50% (“momentum transfer” approximation) at 15 eV incident energy, 13% vs. 22% at 30 eV and 20% vs. 29% at 50 eV. The present approximation did also compare favourably to the previous method in the cases of methane and ethylene [21] and will shortly replace it in our simulation code LEPTS [22].

![Figure 6](image)

**Figure 6.** Inelastic angular distributions obtained with the present approximation. Experimental data for energy losses of: •, 6.45 eV; ■, 7.3 eV; ▲, 8.1 eV; ◇, 9.15 eV; ●, 10.5 eV.

6. Discussion and conclusion

Taking into account the results obtained by our group and complementary data taken from literature, a self-consistent set of recommended data can be put together for the energy range 1 eV - 10 keV. The general strategy followed here is to give priority to experiments and use theoretical data only if no adequate experimental values are available.

Regarding integral cross sections, the different interaction channels are studied separately and then their sum compared to the total CS, which contains the lowest uncertainty and is taken to be accurate. The recommended total scattering CS are the present total scattering CS completed with data from [8] for low energies. Present calculated elastic CS show best agreement with the experiment from Allan [14], which does furthermore cite the best energy resolution, suggesting a lower contamination from rotational excitation processes. Recommended elastic CS are thus based on those two sets of results. The vibrational excitation CS values represent an estimation based on the absolute integral values and differential excitation functions published in [14]. Since no experimental results are available for rotational excitation, values were taken from our calculation and adjusted in absolute magnitude to the remaining CS (σ_{rot} - σ_{el} - σ_{vb}) below the onset of ionization, electronic excitation and neutral dissociation. Electronic excitation CS are based on the Flinders measurements [12], already described, and their extrapolation to higher energies. These data might well fall somewhat below the true value of integral electronic excitation since levels above ~ 8.5 eV are ignored. However, one can argue that this might be approximately compensated for by the fact that some excitations lead to neutral dissociation which is counted separately in its own channel. Recommended ionization CS originate from our experiment and the calculation of Mozejko and Sanche [10]. The remaining CS for energies ≥ 15 eV (σ_{rot} + σ_{el} + σ_{vb} + σ_{ion} - σ_{exc}) are attributed to neutral dissociation not otherwise accounted for. The resulting interaction data set is therefore presented in table 1.

Due to the good agreement between our calculated elastic DCS and the experiments at higher energies, we recommend using IAM-SCAR results for ≥ 30 eV. For lower energies, experimental data by [13,14] should be used for any application. A selection of the resulting recommended elastic angular
distributions, for the whole energy range, is presented in figure 7. The complete table for incident energies in the range 1-10000 eV and angles 0°-180° is available online as supplementary data. For obtaining inelastic angular distributions, the empirical formula proposed in section 5 can be used together with the recommended elastic DCS.

Table 1. Recommended integral cross sections in 10⁻²⁰ m².

| E (eV) | total | elastic | rotation | vibration | electronic excitation | ionisation | neutral dissociation |
|-------|-------|---------|----------|-----------|-----------------------|------------|---------------------|
| 1     | 43.4  | 20.4    | 21.75    | 1.22      | -                     | -          | -                   |
| 1.5   | 42.6  | 26.2    | 15.25    | 1.16      | -                     | -          | -                   |
| 2     | 42.7  | 29.6    | 11.80    | 1.29      | -                     | -          | -                   |
| 3     | 42.2  | 32.2    | 8.25     | 1.76      | -                     | -          | -                   |
| 4     | 42.8  | 33.6    | 6.40     | 2.30      | -                     | -          | -                   |
| 5     | 50.7  | 41.0    | 5.20     | 4.54      | -                     | -          | -                   |
| 7     | 51.3  | 41.7    | 3.86     | 5.74      | -                     | -          | -                   |
| 10    | 49.8  | 41.4    | 2.80     | 5.6       | -                     | -          | -                   |
| 15    | 46.5  | 37.2    | 1.94     | 3.28      | 1.997                 | 1.63       | 0.456               |
| 20    | 46.2  | 31.1    | 1.50     | 1.94      | 5.056                 | 4.38       | 2.18                |
| 30    | 44.4  | 23.0    | 1.03     | 0.901     | 5.563                 | 8.37       | 5.54                |
| 40    | 43.2  | 19.2    | 0.795    | 0.523     | 5.502                 | 10.6       | 6.55                |
| 50    | 41.0  | 16.8    | 0.645    | 0.343     | 4.849                 | 11.2       | 7.16                |
| 70    | 36.5  | 13.9    | 0.475    | 0.182     | 1.977                 | 12.0       | 8.02                |
| 100   | 31.4  | 11.5    | 0.340    | 0.093     | 0.948                 | 12.5       | 6.02                |
| 150   | 26.3  | 9.27    | 0.235    | 0.043     | 0.411                 | 12         | 4.34                |
| 200   | 22.0  | 7.90    | 0.180    | 0.0250    | 0.227                 | 9.89       | 3.78                |
| 300   | 16.6  | 6.19    | 0.125    | 0.0116    | 0.0986                | 7.9        | 2.28                |
| 400   | 13.7  | 5.15    | 0.095    | 0.0067    | 0.0545                | 6.57       | 1.82                |
| 500   | 11.8  | 4.45    | 0.075    | 0.0044    | 0.0344                | 5.64       | 1.60                |
| 700   | 9.08  | 3.53    | 0.055    | -         | 0.0172                | 4.37       | 1.10                |
| 1000  | 6.96  | 2.70    | 0.040    | -         | 0.0083                | 3.39       | 0.822               |
| 1500  | 5.11  | 1.95    | 0.027    | -         | 0.0036                | 2.49       | 0.639               |
| 2000  | 3.90  | 1.54    | 0.021    | -         | -                     | 1.99       | 0.349               |
| 3000  | 2.84  | 1.09    | 0.015    | -         | -                     | 1.46       | 0.276               |
| 4000  | 2.17  | 0.851   | 0.011    | -         | -                     | 1.16       | 0.148               |
| 5000  | 1.78  | 0.700   | 0.009    | -         | -                     | 0.978      | 0.093               |
| 7000  | 1.327 | 0.518   | 0.006    | -         | -                     | 0.750      | 0.052               |
| 10000 | 0.958 | 0.378   | 0.005    | --        | --                    | 0.568      | 0.007               |

Since no significant variation with energy has been observed in our high-energy EEL spectra, a unique, average distribution (figure 3, right panel) is recommended to be used for the whole range of energies studied here. For applications, particularly interested in low energies (≤ 50 eV), that distribution can be adapted by including the excitation detail of the low energy spectra and by forcing a faster decrease after the ionization maximum, as sketched in figure 3 by the dashed line for 30 eV.

Summarizing, here we have used previous and new experimental and theoretical results in order to establish a complete set of electron-THF interaction data in the range 1 eV - 10 keV. This data base includes integral and differential cross sections for elastic and inelastic interactions and an energy loss distribution. Among other applications, it can be used as input parameters for the Low Energy Particle Track Simulation (LEPTS) code [22] for simulating electron transport through THF.
Figure 7. Recommended elastic DCS.

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