

## Near-threshold vibrational excitation and elastic electron scattering from N<sub>2</sub>

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**Abstract.** Angular dependences of the elastic scattering and the vibrational excitation of N<sub>2</sub> have been measured for impact energies between 0.1 and 1.5 eV in the angular range from 15 to 135°. Absolute differential and integrated cross sections are presented. The elastic scattering can be described reasonably well by the modified effective range theory for 0.1 eV. The integral vibrational excitation cross sections are in good agreement with swarm results.

### 1. Introduction

Low-energy electron scattering from N<sub>2</sub> has been investigated experimentally (for example: Schulz 1964, Ehrhardt and Willmann 1967, Engelhardt *et al* 1964, Jung *et al* 1982) as well as theoretically (for example: Chen 1966, Chandra and Temkin 1976a, b, Read 1968, 1972, Read and Andrick 1971) mainly with respect to the <sup>2</sup>Π<sub>g</sub> shape resonance with its oscillatory substructure. No crossed-beam-type measurements in the energy region below 1.2 eV have been performed up to now. Swarm experiments (Engelhardt *et al* 1964) have covered this energy region. Recent measurements by Allan (1985) show good agreement with these swarm data below 1.2 eV, but the comparison of the data is quite indirect because the measured signal is a sum of the counting rates for the scattering angles 0° and 180°. Therefore assumptions have to be made about the angular dependence of the cross sections. The absolute value has been taken from crossed-beam experiments of Jung *et al* (1982). However, especially for the vibrational excitation cross sections, the swarm results yield cross sections which are roughly one order of magnitude lower than the corresponding values obtained by the crossed-beam experiment of Schulz (1964) at 1.2 eV. This discrepancy between different experimental results did not allow a decision on the validity of different theories. Two models for this threshold energy region are under discussion:

(i) pure direct, non-resonant vibrational excitation via the polarisation potential, as described by Breig and Lin (1965); and

(ii) pure resonant scattering, first introduced by Chen (1966).

Obviously, there is a need for experimental differential cross sections (DCS) with more detailed information to clarify the nature of the scattering mechanism. Especially

in the case of  $N_2$  this is a challenging task for an experimentalist since one has to overcome the difficulties of measuring cross sections of the order of some  $10^{-19} \text{ cm}^2$  at primary energies of some tenths of an electronvolt.

## 2. Apparatus

The measurements are performed with the crossed-beam electron spectrometer described in detail in Kochem *et al* (1985). This spectrometer has been improved with respect to the accessible scattering angle region, now reaching from  $-25^\circ$  to  $140^\circ$ . In addition the electron gun has been equipped analogously to the detector with a double  $127^\circ$  cylindrical energy disperser, allowing energy resolution below 20 meV and a more effective suppression of background signals, especially in the small-angle region. This decrease of the background in the forward scattering direction is probably due to the reduction of the divergence angle in the tandem selector system. Thus the filling factor in the following lens system is diminished and the electrons can be better collimated. The background is measured by determining the counting rate without inlet of nitrogen in the scattering centre. To ensure similar conditions of the electron optics, nitrogen is introduced somewhere else into the vacuum chamber just to reach the same pressure in the ionisation gauge. In addition to that, the current on the exit slit of the second selector is controlled, which is generally a good indication of the stability of the electron gun.

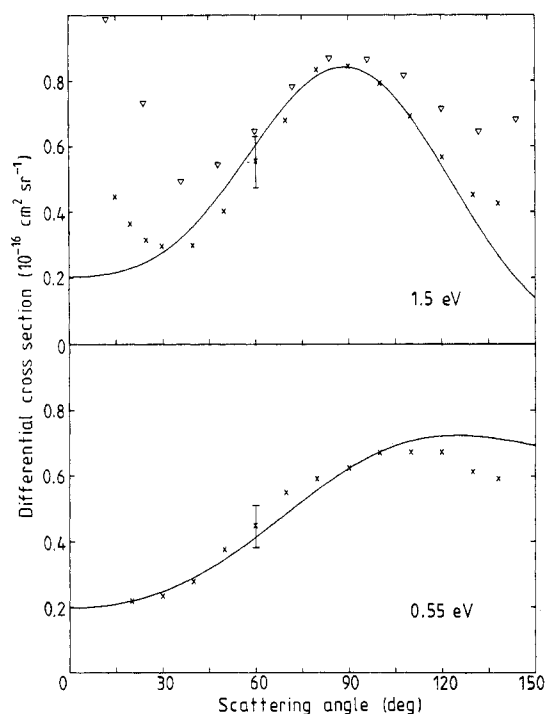
## 3. Results

### 3.1. Elastic scattering

Figure 1 shows the angular dependence of the elastic  $e^- + N_2$  cross sections at 1.5 and 0.55 eV. At 1.5 eV collision energy the scattering is symmetrical around  $90^\circ$  in agreement with the measurements of Ehrhardt and Willmann (1967) and Shyn and Carignan (1980). The increase in the cross section for scattering angles smaller than  $30^\circ$  observed by Shyn and Carignan (1980) can be confirmed qualitatively. In the present measurements the increase is even sharper than in the results of Shyn and Carignan. This behaviour has not been predicted by the calculations of Siegel *et al* (1978).

The full curve in figure 1 represents the result of calculations of Morrison and Collins (1978). From their partial cross sections, phaseshifts  $\delta_{lA}$ , which have been listed in table 1, could be obtained. In general the partial cross sections had to be interpolated to match the experimental primary energies. With these phases (s-d and s-p interference is taken into account, but no p-d interference terms) the determination of the differential cross section (DCS) is straightforward. However, the resulting absolute values are generally higher than our experimental results (25% at 0.55 eV and 47% at 1.5 eV). Therefore, they are presented in figure 1 normalised to the experiment at  $90^\circ$  scattering angle to show the good agreement in shape.

For lower collision energies, backscattering becomes more and more prominent, resulting mainly from the interference of the  $s\sigma$  and  $p\pi$  partial waves (see table 1). This tendency is in agreement with the calculations of Siegel *et al* (1978) and Morrison and Collins (1978); comparison with experimental data is only possible via the total

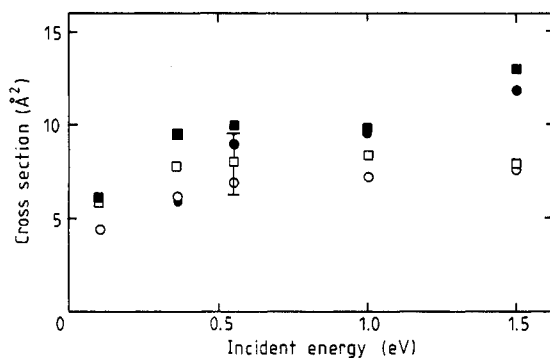


**Figure 1.** Angular dependence of the elastic differential cross section (DCS) at 1.5 and 0.55 eV impact energy:  $\times$ , this work;  $\nabla$ , Shyn and Carignan (1980); —, calculation of Morrison and Collins (1978).

**Table 1.** Phaseshifts  $\delta_{l\Lambda}$  extracted from the work of Morrison and Collins (1978). The phase convention was to set  $\delta_{20}$  identically to zero.

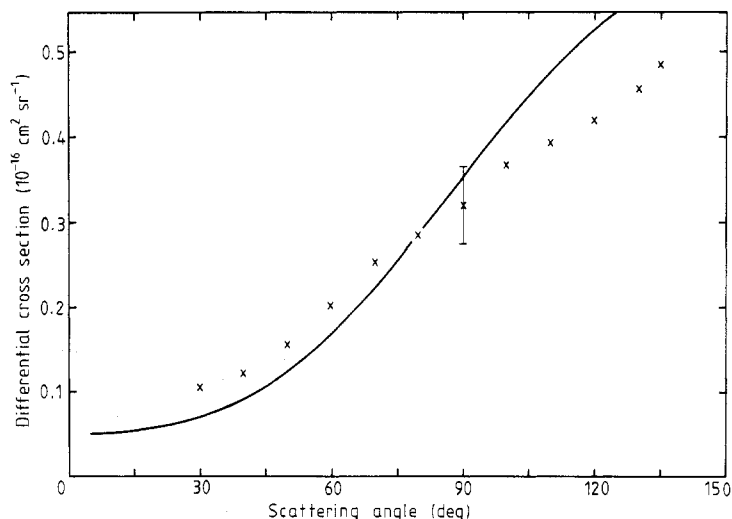
$E_0$ (eV)	$\delta_{l\Lambda}$ (rad)				
	$\delta_{00}$	$\delta_{10}$	$\delta_{11}$	$\delta_{21}$	$\delta_{22}$
0.1	-0.115	-0.003	0.030	0.002	0.009
0.35	-0.275	-0.008	0.056	0.012	0.020
0.55	-0.362	-0.016	0.064	0.022	0.032
1.0	-0.513	-0.051	0.057	0.063	0.054
1.5	-0.640	-0.100	0.030	0.180	0.079

cross section (TCS). As the contributions from vibrational excitation to the TCS can be neglected (see § 2), the TCS can be obtained by a suitable extrapolation of the elastic DCS towards  $0^\circ$  and  $180^\circ$  respectively, followed by numerical integration. The TCS obtained in this way agree within the error bars with those of Baldwin (1974), but are systematically lower (see figure 2). Also shown in figure 2 are the momentum transfer cross sections (MTC) of Engelhardt *et al* (1964) in comparison with the corresponding results of this work. Again, rotational excitation has been assumed not to contribute to the elastic peak. Excellent agreement is found for 0.1 eV collision energy for the MTC. The TCS is found to be smaller than the MTC, which reflects the dominant backscattering behaviour in this energy region.



**Figure 2.** Energy dependence of the total cross section (TCS) and the momentum transfer cross section (MTC):  $\circ$ , TCS, this work;  $\square$ , MTC, this work;  $\bullet$ , TCS, Baldwin (1974);  $\blacksquare$ , MTC, Engelhardt *et al* (1964).

Figure 3 shows the angular dependence of the elastic DCS at a collision energy of 0.1 eV. For comparison the results of the modified effective range theory (MERT) are shown as the full curve. The Morrison and Collins (1978) results are not shown separately as they nearly coincide with the MERT. Using the known molecular properties  $\alpha_0=11.9$  au,  $\alpha_2=4.2$  au,  $Q=-1.13$  au and  $A=0.44$  au for the isotropic and anisotropic polarisability, the quadrupole moment and the scattering length, respectively, theory and experiment agree within 25% over the whole measured isotropic and anisotropic polarisability, the quadrupole moment and the scattering length, respectively, theory and experiment agree within 25% over the whole measured angular range. It should be noted that rotational excitation with  $\Delta j=\pm 2$  has been accounted for by the use of the Born quadrupole approximation (Takayanagi and Itikawa 1970). For higher energies MERT fails to explain the experimental results. This also holds if one calculates the total cross sections with the MERT formula given by Chang (1981), yielding a value



**Figure 3.** Angular dependence of the elastic DCS at 0.1 eV collision energy:  $\times$ , present result; —, MERT.

of 20.2 Å<sup>2</sup> for 0.35 eV primary energy. This value is too large by more than a factor of 2 in comparison with experiment. Finally, the elastic DCS for all measured energies are given in table 2.

### 3.2. Vibrational excitation

Vibrational excitation in N<sub>2</sub> is dominated by the <sup>2</sup>Π<sub>g</sub> shape resonance centred at about 2.4 eV. There the total vibrational cross section (sum of the integral cross sections for transitions to all different final vibrational states) exceeds the low-energy off-resonance cross section by more than two orders of magnitude (Engelhardt *et al* 1964). However, as mentioned above, the nature of the excitation mechanism in the vibrational threshold region below the resonance has not yet become clear.

**Table 2.** Elastic DCS for all measured energies (in units of 10<sup>-16</sup> cm<sup>2</sup>).

Scattering angle (deg)		Collision energy (eV)				
		0.1	0.35	0.55	1.0	1.5
11082	10	—	—	—	0.121	—
11083	20	—	0.121	0.221	0.149	0.365
	30	0.106	0.133	0.255	0.205	0.297
	40	0.123	0.164	0.280	0.277	0.299
	50	0.157	0.223	0.377	0.393	0.403
	60	0.203	0.301	0.449	0.522	0.554
	70	0.254	0.379	0.551	0.625	0.681
	80	0.286	0.456	0.592	0.702	0.834
	90	0.322	0.520	0.626	0.722	0.846
	100	0.369	0.564	0.670	0.753	0.791
	110	0.396	0.638	0.674	0.727	0.692
	120	0.422	0.692	0.674	0.689	0.567
	130	0.459	0.724	0.614	0.632	0.452
	138	0.488	0.746	0.593	0.615	0.424
	σ <sub>int</sub>	4.44	6.15	6.81	7.30	7.43

Figure 4 shows the angular dependence of the vibrational excitation cross section for  $v=0$  to 1 at a collision energy of 1.5 eV. The influence of the <sup>2</sup>Π<sub>g</sub> resonance is obviously quite strong, but the angular behaviour cannot be purely due to this resonance: the ratio of the DCS at 20° to that at 90° should be 2.2 for pure resonant scattering, whereas the measured ratio exceeds a factor of 3. Following Chang (1985) this discrepancy may be partly due to an s-d interference term which gives rise to a contribution proportional to  $(1 - 6 \cos^2 \theta + 9 \cos^4 \theta)$  to the pure resonant DCS, resulting in a slight enlargement of the considered ratio. The overall agreement however could not be improved.

If direct scattering contributions are responsible for this discrepancy, their angular dependence should be forward-peaked when they are, for the sake of simplicity, assumed to add incoherently.

As the N<sub>2</sub> vibration is not infrared-active, the interactions that may be responsible for the excitation process are the quadrupole and polarisation potential. The quadrupole and anisotropic polarisability transitions lead to isotropic and slightly backward-peaked angular distributions, respectively, if calculated in the Born approximation

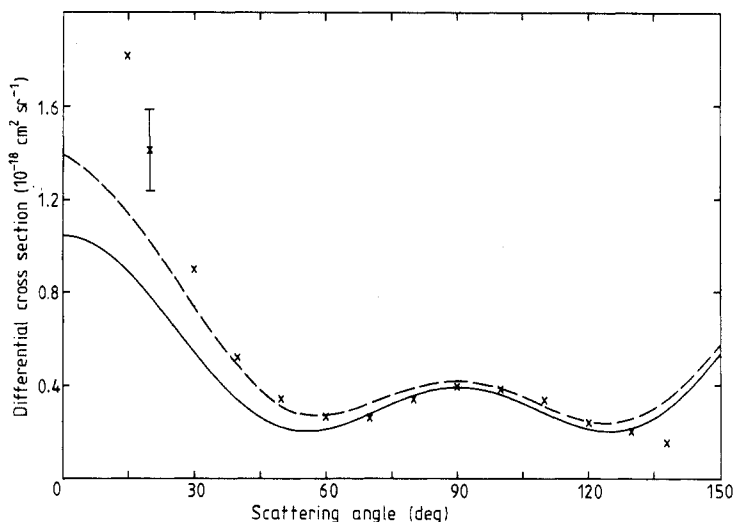


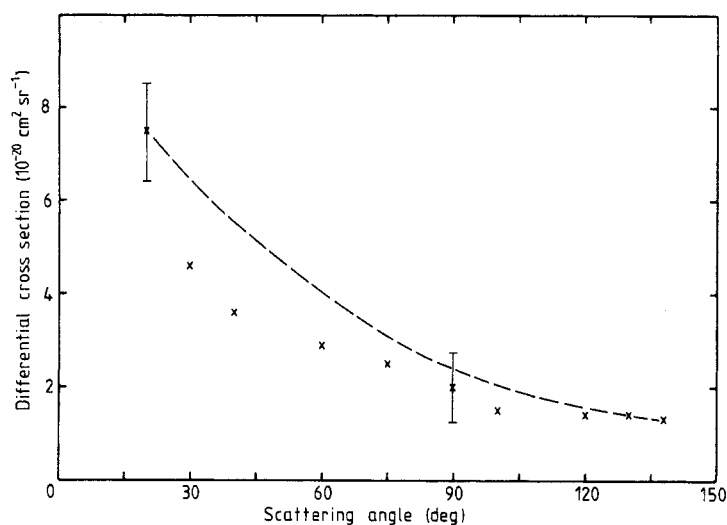
Figure 4. Angular dependence for vibrational excitation of  $v = 1$  level at 1.5 eV:  $\times$ , present result; ---, incoherent sum of resonant and direct excitation; —, pure resonant scattering.

(Takayanagi and Itikawa 1970, Breig and Lin 1965). The deviations from pure resonant scattering cannot be explained by the above mechanisms. In contrast, isotropic polarisability transitions yield a forward-peaked angular behaviour with both the absolute cross sections and the shape of the angular dependence being rather sensitive to the cutoff of the  $1/r^4$  potential for small values of  $r$ . With the transition matrix element of 0.344 au given by Breig and Lin (1965), a satisfactory fit can be obtained for a cutoff radius  $r_c$  of  $3.1a_0$ , when a  $1 - \exp[-(r/r_c)^6]$  cutoff function is used. So the angular dependence shown in figure 4 can be explained by the incoherent sum of resonant and direct scattering.

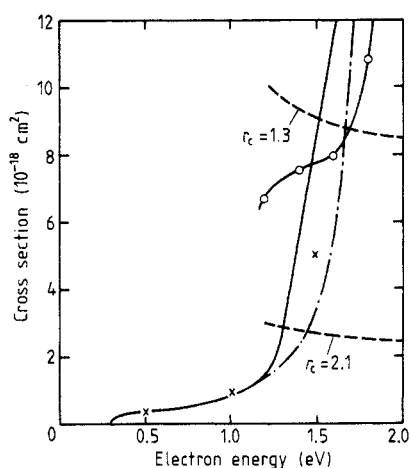
The Born polarisation approximation, however, fails to fit the angular dependence for vibrational excitation near threshold at 0.5 eV plotted in figure 5 if the same cutoff radius is used. Neglecting resonant scattering, reasonable agreement can be achieved for this primary energy with a cutoff radius of  $4.5a_0$  for the direct isotropic polarisation contribution.

Generally, the selection rule for transitions caused by the polarisation potential allows  $\Delta J = 0$  ( $J$  being the total angular momentum) for the target molecule; therefore pure s-wave scattering of the electron is possible connected with a deep penetration into the interaction potential because of the lack of a centrifugal barrier. But this restricts the quantitative applicability of this approximation because the potential in the atomic core region is not described correctly by the terms that are used to analyse the asymptotics of the potential.

In figure 6 integral cross sections are compared, which have been obtained by a swarm data analysis of Engelhardt *et al* (1964), the crossed-beam data of Schulz (1964) and this work. For 0.5 eV and 1 eV primary energy, the swarm data and the results of this work are in excellent agreement, whereas for 1.5 eV there is a deviation of about a factor of 2. The broken curves in figure 6 give the Born result calculated by Breig and Lin (1965). Again, the failure to describe the actual physical situation by the Born polarisation approximation with a fixed cutoff radius becomes obvious. The measured DCS for vibrational excitation are finally summarised in table 3.



**Figure 5.** DCS for vibrational excitation of  $v=1$  level at 0.5 eV:  $\times$ , present result; ---, Born approximation with  $r_c = 4.5a_0$ .



**Figure 6.** Integral cross sections for vibrational excitation (see Lane 1980):  $\times$ , this work;  $\circ$ , Schulz (1964); ---, Breig and Lin (1965); - · -, Chen (1966); —, Engelhardt *et al* (1964).

For the interpretation of the measured data both the model of pure resonant vibrational excitation down to threshold (Chen 1966) and pure direct scattering (Breig and Lin 1965) do not hold. The data show that between about 1 eV and 1.5 eV both interaction mechanisms have to be taken into account whereas in the near-threshold energy region the resonance seems to have no influence. This statement is also supported by the fact that in the threshold region the  $v=0 \rightarrow 2$  excitation probability is small in comparison to the  $v=0 \rightarrow 1$  excitation. Resonant interaction in general raises the probability to excite higher harmonics.

**Table 3.** DCS for vibrational excitation at 0.5, 1.0 and 1.5 eV (in units of  $10^{-18} \text{ cm}^2$ ).

$\theta$ (deg)	$E$ (eV)		
	0.5	1.0	1.5
15	—	—	1.82
20	0.075	0.178	1.41
30	0.046	0.176	0.90
40	0.036	0.136	0.519
50	—	0.105	0.342
60	0.029	0.093	0.265
70	—	0.083	0.259
80	—	0.065	0.339
90	0.020	0.057	0.395
100	0.015	0.043	0.381
110	—	0.040	0.334
120	0.014	0.026	0.239
130	0.014	0.033	0.199
138	0.013	0.046	0.154
$\sigma_{\text{int}}$	0.48	0.93	5.2

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## References

- Allan M 1985 *J. Phys. B: At. Mol. Phys.* **18** 4511  
 Baldwin G C 1974 *Phys. Rev. A* **9** 1225  
 Breig E L and Lin C C 1965 *J. Chem. Phys.* **43** 3839  
 Chandra N and Temkin A 1976a *Phys. Rev. A* **13** 138  
 — 1976b *Phys. Rev. A* **14** 507  
 Chang E S 1981 *J. Phys. B: At. Mol. Phys.* **14** 893  
 — 1985 Private communication  
 Chen J C Y 1966 *Phys. Rev.* **146** 61  
 Ehrhardt H and Willmann K 1967 *Z. Phys.* **204** 462  
 Engelhardt A G, Phelps A V and Risk C R 1964 *Phys. Rev.* **135** A1566  
 Jung K, Antoni Th, Müller R, Kochem K-H and Ehrhardt H 1982 *J. Phys. B: At. Mol. Phys.* **15** 3535  
 Kochem K-H, Sohn W, Jung K, Ehrhardt H and Chang E S 1985 *J. Phys. B: At. Mol. Phys.* **18** 1253  
 Morrison M A and Collins L A 1978 *Phys. Rev. A* **17** 918  
 Lane N F 1980 *Rev. Mod. Phys.* **52** 29  
 Read F H 1968 *J. Phys. B: At. Mol. Phys.* **1** 893  
 — 1972 *J. Phys. B: At. Mol. Phys.* **5** 255  
 Read F H and Andrick D 1971 *J. Phys. B: At. Mol. Phys.* **4** 911  
 Schulz G J 1964 *Phys. Rev.* **135** A998  
 Shyn T W and Carignan G R 1980 *Phys. Rev. A* **22** 923  
 Siegel J, Dill D and Dehmer J L 1978 *Phys. Rev. A* **17** 2106  
 Takayanagi K and Itikawa Y 1970 *Advances in Atomic and Molecular Physics* vol 6 (New York: Academic Press) p 105