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Electron Attachment to Nitric Oxide (NO) Controversy

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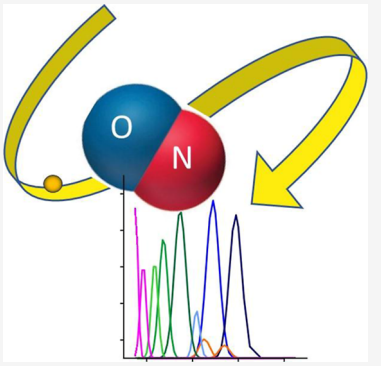
ABSTRACT: We report novel total electron scattering cross sections (TCS) from nitric oxide (NO) in the impact energy range from 1 to 15 eV by using a magnetically confined electron transmission apparatus. The accuracy of the data to within 5% and its consistency across the energy range investigated, shows significant discrepancies from previous works as to the major resonance features and magnitude of the TCS. Within the shape of the TCS, we have identified nine features which have been assigned to electron attachment resonances, most of them reported for the first time, while a comprehensive analysis of those peaking at 7.0, 7.8, and 8.8 eV has led to solve the controversy about dissociative electron attachment (DEA) cross-section that persisted for more than 50 years.



1. INTRODUCTION

Nitric oxide (NO) is a very reactive molecule frequently interacting in biological and environmental media. In order to understand the underlying molecular mechanisms related to electron induced processes within these reactions, electron affinities of NO and resonant formation of the NO^- anion (see Alle et al.[1] and references therein) have been experimentally analyzed. Also, calculations with the R-matrix method[2] and with the Schwinger multichannel method[3] evidenced the presence of resonances superimposed on the elastic scattering cross sections. These studies are mainly focused on the elastic and vibrationally excited resonances of the lowest-lying electronic state of the anion. In particular, Trevisan et al.[4] calculated the contribution of these resonances to the elastic and vibrationally inelastic cross section in the 0−2 eV energy range as well as the corresponding dissociative electron attachment (DEA) cross-section yielding O^- (^2P) in the ground state. For higher energies, Rapp and Briglia[5] (1965) measured the total anion current generation from 6.5 to 13 eV, where a broad double peak structure attributed to DEA processes has been reported. Note that these are the only direct absolute negative ion formation cross-section values available in the literature to date. The previous mass spectrometric analysis of Hagstrum and Tate[6] (1941) concluded that O^- was the only anion formed by DEA to NO. The onset potential obtained for the O^- formation resonant process at 7.0 ± 0.3 eV, is higher than that obtained by Hanson (1937)[7] by extrapolating to zero the kinetic energy of the detected anions. Additional measurements from Chantry[8] (1968) provided the total anion current together with mass analysis and kinetic energy of the formed species. Again, only O^- fragments were detected with an energy threshold of 7.5 ± 0.1 eV given by the linear fit of the O^- kinetic energy distribution. Considering the available excited states of the neutral nitrogen atom, the energy thresholds (E_{th}) of the possible DEA processes are

$$e^- + NO \rightarrow [NO^-]^* \rightarrow O^-(^2P) + N^*(^4S) \quad (E_{th} = 5.07\ eV) \tag{1a}$$

$$e^- + NO \rightarrow [NO^-]^* \rightarrow O^-(^2P) + N^*(^2D) \quad (E_{th} = 7.46\ eV) \tag{1b}$$

$$e^- + NO \rightarrow [NO^-]^* \rightarrow O^-(^2P) + N^*(^2P) \quad (E_{th} = 8.66\ eV) \tag{1c}$$

with $[NO^-]^*$ the temporary negative ion (TNI). By comparing the energy thresholds calculated from the bond dissociation energy, electron affinity and the possible excitation energies of the remaining nitrogen atom with the corresponding experimental energy threshold, and considering the slope of the detected O^- current as a function of the electron impact current (see ref 8 for details), Chantry concluded that the only process leading to N* in the ^2D excited state (1b) can contribute to the observed O^- fragment.

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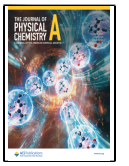

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Further studies in this energy range provided relative or even absolute values of the dissociative attachment cross sections[9] through normalizing procedures or using the relative flow technique (see ref 9 and references therein). In spite of the experimental effort made, important discrepancies about the energy threshold and the intensity of the resonances leading to O^- formation persisted. Some years later Orient and Chutjian,[10] using combined electric and magnetic spectrometry techniques, concluded that the three channels (1a−1c) contribute to the broad O^- feature reported in refs 4 and 7, with reaction (1a) being clearly dominant and yet assigning cross section-values to all of them. This additional controversy motivated new studies based on more accurate techniques, using high resolution electron beams and time-of-flight mass spectrometry. Chu et al.[11] showed that Orient and Chutjian's conclusions were possibly wrong and stated that the dissociative attachment process to NO is dominated by reaction (1b) with a small contribution (15−20%) of reaction (1c). This conclusion was also supported by the mass spectrometry analysis of Denifl et al.[12] and later commented by Illenberger and Märk.[13] Nonetheless, Orient and Chutjian[14] replied to this comment suggesting that discrepancies can arise from the different angles of detection and claiming that their experimental system has the ability to collect ions ejected in all directions. From the persisted ambiguities within the scientific community, Allan[15] used a high-resolution spectroscopy technique to conclude that only signals corresponding to reaction (1b) yield a DEA process. In the conditions of Allan's experiment, without external magnetic fields, no signals arising from reactions (1a) and (1c) were detected. Thus, from the different rationales put forward, and after all the above discussions it seems that Chantry's interpretation was right and only one channel (1b) contributes to the anion generation by DEA to NO. Despite the considerable information, including higher energy Feshbach resonances,[16] some doubts about the influence of specific experimental conditions on the results persist and relevant inconsistences in the absolute value of the DEA cross sections require further attention. More recently, Nandi et al.[17] combined accurate experimental techniques (viz. ion momentum imaging) with R-matrix scattering calculations with the aim of definitively clarifying the above controversy. From the experimental data they concluded that, in disagreement with Allan's electron spectroscopic analysis, O^- detected signals can result from reactions (1b) and (1c).

In addition to previous reports on electron scattering by NO molecules,[18,19] in 2019 Song et al.[20] reviewed the current situation of electron scattering cross sections from NO, N_2O and NO_2. As derived from this study, no direct electron attachment data are available in the literature. Only the aforementioned contradictory DEA results are reported. As shown by Alle et al.[1] for the lower energies, electron attachment (EA) resonances appear as relative maxima of the total cross-section (TCS) values. However, apart from those low-energy results,[1] the TCS measurements reported by Song et al.[20] do not show any resonance signature above 2 eV (see Figure 2 of ref 20). These measurements were performed by Zecca et al.,[21] Dalba et al.[22] and Szmytkowski and Maciag[23] and, considering the precedent discussion about electron attachment resonances leading to O^- formation processes around 8 eV, a new inconsistency appeared in the electron-NO scattering literature. This contradiction was not discussed by Song et al.[20] and is probably due to deficiencies in the energy resolution used in these experiments, but it is clear that still requires experimental verification.

The above considerations, together with the recent interest in nitro-compounds as potential radiosensitizers[24−26] via NO and NO_2 generation and the relevance of these reactive species in the biosphere conditions[27] motivated the present study.

2. EXPERIMENTAL METHOD

Here we use a magnetically confined electron transmission apparatus to determine the total electron scattering cross sections from NO, for electron impact energies ranging from 1 to 15 eV, with a total uncertainty limit of about 5%. The considered energy range included all the expected electron attachment resonances. Details on the experimental setup and procedure can be found in a previous publication.[28] The quoted energy spread of the magnetically confined electron beam is better than 200 meV but the cutoff procedure followed to measure the transmitted beam intensity provided an effective energy resolution of about 50 meV. Details on the uncertainty estimation procedure and a deep analysis of possible error sources, including systematic errors, can be found in ref 28. Basically, the total ±5% assigned to the absolute TCS values is the result of adding in quadrature the statistical uncertainties (4%), absolute pressure and temperature determination (1%) and fitting procedures (1%). The present results are not corrected for the systematic error due to electrons elastically scattered in the acceptance angle of the detector.[28] This fact does not affect to the discussion and conclusions of this study. Details on detector efficiency and attenuation curve analysis can also be found in ref 28.

As already mentioned, if the energy resolution of the electron scattering experiment is good enough, resonances should appear as a clear increase of the TCS around the resonant energy. By assuming that the continuum background cross section over which the resonances appear is mainly formed by elastic and electronic excitation processes, we have subtracted from our experimental TCS values the cross sections, corresponding to these processes, recommended by Song et al.[20] The resultant cross sections have been assigned to EA processes and the uncertainty of their absolute values has been estimated to be about 15% by adding in quadrature the uncertainty limits of the cross sections intervening in the subtraction procedure. The resulting resonances have been analyzed through a standard Gaussian fit by using a commercial software (MagicPlotStudent). These resonances are characterized by their respective widths, which are the convolution of the energy resolution (instrumental width) with their vibrational structure and limited by the lifetime of the transient negative ion formed during the collision process, and thresholds (here assigned to the energy at which the cross section is 10% of that at the peak of the Gaussian curve). Both maximum and threshold values assigned to these resonances are affected by the estimated ±15% uncertainty limit.

3. RESULTS AND DISCUSSION

The results of the present TCS measurements, with total uncertainties within 5%, are shown in Figure 1 and the corresponding numerical values are listed in Table 1. A detailed description of all the error sources and the analysis of the total uncertainty limits can be found in ref 28.

Figure 1 reveals that the TCS features we find between 3 and 10 eV are not accounted by the TCS values recently

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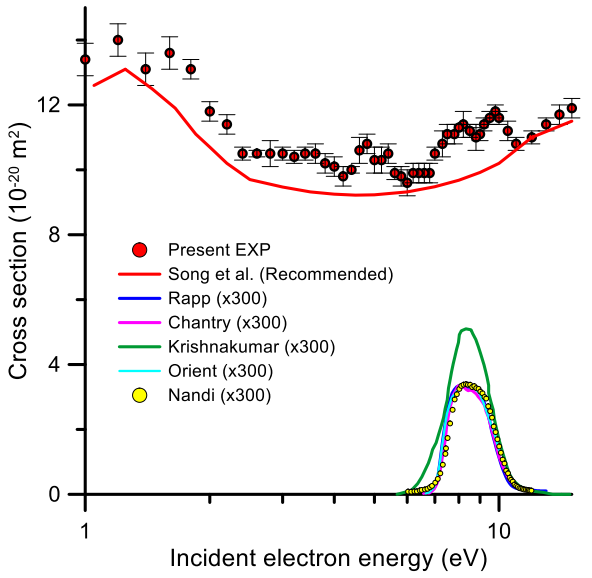

Figure 1. Total electron scattering cross sections (red circle, present experimental results; red emdash, recommended values from ref 20) and O^- production by DEA cross sections (indigo emdash, from ref 5; pink emdash, from ref 8: green emdash, from ref 9; blue emdash, from ref 10; yellow circle, from ref 17).

recommended by Song. et al.[20] As mentioned above these peaks on the TCS values correspond to the contribution of resonant EA processes. The TCS values corresponding to the minima of these features are in good agreement with the recommended data but these features were not detected in previous experiments. We have also plotted in this figure the O^- production by DEA cross sections discussed above. Such DEA resonances are within the prominent peaks we measured between 7 and 12 eV. Other relevant aspect is the absolute value assigned to the DEA cross-section is about 2 orders of magnitude lower than that corresponding to the EA peak.

For a deeper analysis of the experimental findings we have plotted in Figure 2 the EA cross-section values resulting from subtracting of the Song et al.[20] recommended elastic plus electronic excitation cross sections from our TCS experimental values. The resulting EA cross sections are also listed in Table 1 where the numbers used in the subtraction procedure are the difference between columns 2 and 3 of this table. As seen in Figure 2, a significant number of resonances appear between 1 and 12 eV, reaching peak values of about 1.7×10^{-20} m^2. The intense peak at 7.8 ± 0.2 eV agrees in shape with that of O^- production previously reported.[6,8] The threshold energy of this resonance, defined as the energy value at which it reaches the 10% of its maximum value, is about 6.5 eV, which is consistent with that corresponding to reaction (1b), thus leading to O^- formation and leaving the neutral nitrogen atom in the ^2D state. However, its absolute value is 100 times higher than those reported in refs 4 and 8. In both sides of this resonance feature we note two other prominent peaks with threshold energies of 5.5 and 8.7 eV. Note that these energies are very close to the threshold of reactions (1a) and (1c) DEA processes. These results evidence that O^- anions are, in principle, formed via reactions (1a), (1b) and (1c) but the low anion production yield possibly associated with (1a) and (1c) reduces the probability of being detected within the sensitivity limits of the different experimental systems.

Table 1. Total Electron Scattering and Electron Attachment (EA) Cross Sections in 10^{-20} m^2 units with Their Respective Uncertainties (in Brackets)

energy (eV)	total cross section (±5%)	EA cross section (±15%)
1.0	13.4	0.97
1.2	14.0	1.7
1.4	13.1	0.79
1.6	13.6	0.99
1.8	13.1	1.0
2.0	11.8	0.47
2.2	11.4	0.11
2.4	10.5	0.28
2.6	10.5	1.3
2.8	10.5	1.3
3.0	10.5	1.3
3.2	10.4	1.2
3.4	10.5	1.3
3.6	10.5	1.4
3.8	10.2	1.1
4.0	10.1	0.99
4.2	9.8	0.71
4.4	10.0	0.89
4.6	10.6	1.5
4.8	10.8	1.7
5.0	10.3	1.1
5.2	10.3	1.1
5.4	10.5	1.3
5.6	9.9	0.71
5.8	9.8	0.60
6.0	9.6	0.39
6.2	9.9	0.68
6.4	9.9	0.67
6.6	9.9	0.66
6.8	9.9	0.62
7.0	10.5	1.2
7.3	10.8	1.4
7.5	11.1	1.7
7.8	11.1	1.5
8.0	11.3	1.3
8.2	11.4	1.2
8.5	11.2	1.0
8.8	11.0	0.78
9.0	11.1	0.93
9.2	11.4	1.2
9.5	11.6	1.4
9.8	11.8	1.6
10.0	11.6	1.4
10.5	11.2	0.79
11.0	10.8	0.21
12.0	11.0	0.05
13.0	11.4	<0.05
14.0	11.7	<0.05
15.0	11.9	<0.05

Moreover, there are other channels compatible with the electron attachment to NO, i.e.

$$e^- + NO \rightarrow [NO^-]^* \rightarrow e^- + NO^* \qquad (E_{th} = <1\ eV) \tag{1d}$$

$$e^- + NO \rightarrow [NO^-]^* \rightarrow O(^3P) + N^-(^3P) \rightarrow e^- + O(^3P) + N \qquad (E_{th} = 6.57 eV) \tag{1e}$$

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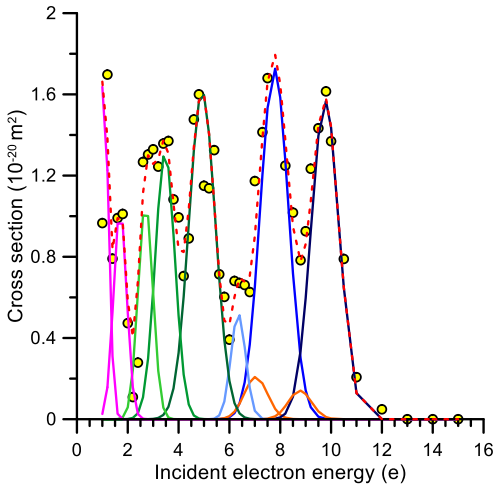

Figure 2. Electron attachment cross sections: circle open, present results (see text for details); red hyphen, Gaussian fit to the present experimental data.

$$e^- + NO \rightarrow [NO^-]^* \rightarrow O(^3P) + N^-(^1D) \rightarrow e^- + O(^3P) + N \qquad (E_{th} = 7.94\ eV) \tag{1f}$$

$$e^- + NO \rightarrow [NO^-]^* \rightarrow O(^1D) + N^-(^3P) \rightarrow e^- + O(^1D) + N \qquad (E_{th} = 8.53\ eV) \tag{1g}$$

Where the energy thresholds of reactions (1e), (1f) and (1g) are taken from ref 17. Reaction (1d) is related to those resonances reported by Alle at al.,[1] which were interpreted by Tennyson and Noble[2] and Trevisan et al.[4] (see these references for details). The resonances we find below 2 eV agree well with these, although our minimum energy limit (1 eV) does not allow to reproduce all the vibrational structure associated with such resonances. The weak resonances shown in Figure 2 at 7.0 and 8.8 eV, with threshold energies around 6 and 8 eV, can be associated with reaction (1e) and (1f). It is well-known that electrons autodetach from N^- anions very fast and the negative nitrogen atom has never been detected in DEA experiments.[17] However, the formation of N^- in DEA to NO has been observed indirectly by detecting the corresponding autodetached electron,[29] while the (1g) process is not distinguishable within the present experimental conditions.

As can be seen in Figure 2, between 2 and 6 eV we found three peaks at 2.8, 3.6, and 5.0 eV, respectively, which have not been previously reported. The threshold energies of these resonances are much lower than those corresponding to the processes represented by (1e), (1f) and (1g) and, since no anion fragments have been detected in previous studies (see ref 17 and references therein), we should assume that they arise from EA processes as those represented by reaction (1d). A proper identification of states intervening in these processes would require an accurate molecular structure calculation including all the intervening states which is out of the scope of this study. However, some hints can be found in previous studies. Sanche and Schulz,[30] using an electron transmission spectrometer, reported some vibrational structures in the range 5−7.5 eV which were identified as two Rydberg electrons attached to the NO^+ ion core. Gresteau et al.[16] studied the excitation function of different vibrational levels of the NO ground state for impact energies from 5 to 7.5 eV. They found resonance positions in agreement with those of Sanche and Schulz[30] for $v = 0$ at 5.0, 5.4, 5.5, and 6.4 eV, along with other resonances which correspond to higher (v = 1, 2, 3 and 4) vibrational numbers (see ref 16 for details). With respect to the resonances we found in the energy range of 2−4 eV, Nandi et al.[17] showed that the $1^1\Delta$ single particle resonance crosses the Franck−Condon region between 2 and 4 eV, which can explain one of these observed features (see ref 17 for details). All these results are consistent with the present resonance composition shown in Figure 2.

4. CONCLUSIONS

In conclusion, using a magnetically confined electron transmission apparatus we have measured the total electron scattering cross section for electron impact energies ranging from 1 to 15 eV. The present energy resolution (<50 meV) has allowed to detect different electron attachment resonances occurring in this energy range. We have identified the three EA processes leading to O^- formation at the energy thresholds predicted by theory (5.0, 7.4, and 8.6 eV) for the corresponding states of the neutral N atom (^{4}S, ^{2}D and ^{2}P, respectively). Since previous DEA measurement only detected O^- formation from the ^{2}D channel (7.4 eV threshold energy), and possibly from the ^{2}P channel (8.7 eV threshold energy), we have concluded that the electron autodetachment process is dominant in these transitions and the O^- detection from the other two channels strongly depends on the sensitivity limits of the different experimental systems. Transitions corresponding to N^- formation have also been detected. These were identified to contribute to the DEA in N-channel, which might not be detected in the DEA experiments based on their experimental timing conditions. Absolute EA cross sections have been derived by combining the present TCS measurements with the recommended integral elastic and electronic excitation cross sections. The present EA cross sections resulted to be 2 orders of magnitude higher than the available DEA cross sections, thus indicating that electron autodetachment is the most probable decay channel of the transient anion formed by electron attachment to NO. We consider that these findings contribute to resolve the dissociative electron attachment to NO controversy existing from 1965 and add accurate total electron scattering and electron attachment cross sections to the available databases which, as a consequence of this study, need to be updated. In order to characterize the new EA resonances reported in this experimental study, further accurate calculations including high energy electronic excited states of the NO^- anion are needed.

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Notes

The authors declare no competing financial interest.

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