PHOTOIONISATION OF ATOMIC OXYGEN AND ATOMIC NITROGEN

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I. Introductory Remarks 1
II. Photoionisation of Atomic Oxygen 2
A. Ground State 2
B. Excited States 7
III. Photoionisation of Atomic Nitrogen 10
A. Ground State 10
B. Excited States 15
IV. Conclusion 16
References 17

I. Introductory Remarks

Photoionisation of atomic oxygen by extreme ultraviolet radiation from the sun is the primary process for the production of energetic electrons and ions in the daytime thermosphere, and photoelectrons from oxygen provide the major source of electron heating in the daytime mid-latitude ionosphere. Atomic oxygen and nitrogen also play important roles in ionisation balance in HII regions, stellar atmospheres and planetary nebulae. For over 50 years therefore knowledge of accurate cross sections for photoionisation of these elements has been sought. In this chapter we review the progress made for total cross sections. (Attention is drawn to an excellent article on ground state oxygen by Seaton, 1987.)

Before commencing, however, it is worth remarking and demonstrating the outstanding achievements of the earliest calculations. The first calculation of the cross section for photoionisation of atomic oxygen was made by Bates *et al.* (1939) using a Hartree self-consistent field approximation without exchange. Bates (1939) then calculated cross sections for all elements from boron to neon using the free-electron wave functions calculated in the field of O^+. These results were improved upon by Bates and Seaton (1949), who employed the Hartree–Fock approximation to evaluate the threshold cross section; the cross section as a function of energy was obtained by normalising

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the results from the general formula of Bates (1946) based upon an approximation which used Slater-type orbitals for bound electrons and Coulomb functions for ejected electrons. Figures 1 and 2 compare these results for photoionisation of the ground state of oxygen and nitrogen with the most recent theoretical calculation and the latest experimental data. Whereas the older calculations were incapable of accounting for resonances, the agreement with the most recent results, in both shape and in magnitude, is truly remarkable.

II. Photoionisation of Atomic Oxygen

A. Ground State

The early calculations considered only ejection of the 2p electron from the $2s^2 2p^4\ ^3P$ ground state of oxygen. Dalgarno and Parkinson (1960) included contributions from the process in which a 2s electron is ejected. In 1964, Dalgarno *et al.* employed the Hartree–Fock formulation to calculate cross

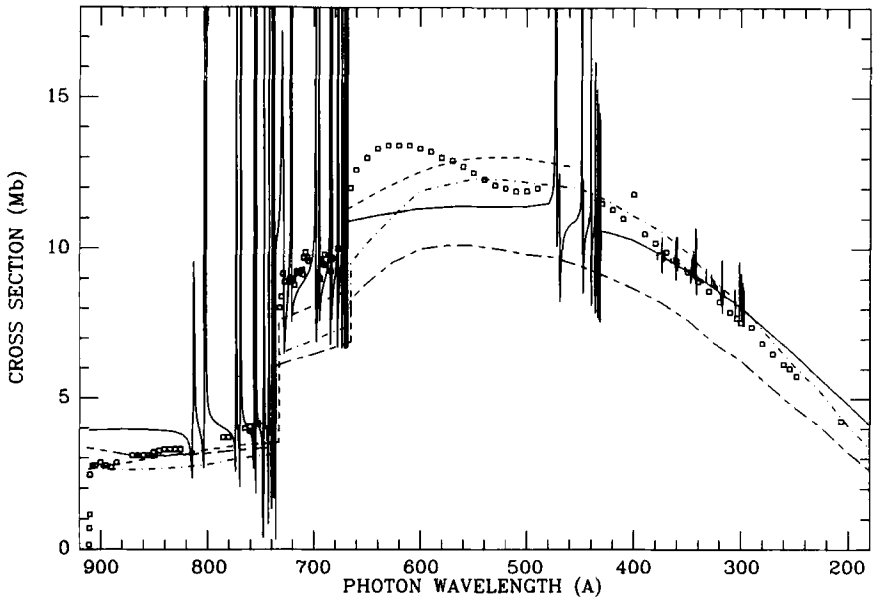

FIG. 1. Photoionisation cross section of oxygen in the ground state. Theory: —— Bell *et al.* (1989); - - - Bates and Seaton (1949); -·-, —, Dalgarno *et al.* (1964) (length and velocity values). Experiment: □ Angel and Samson (1988).

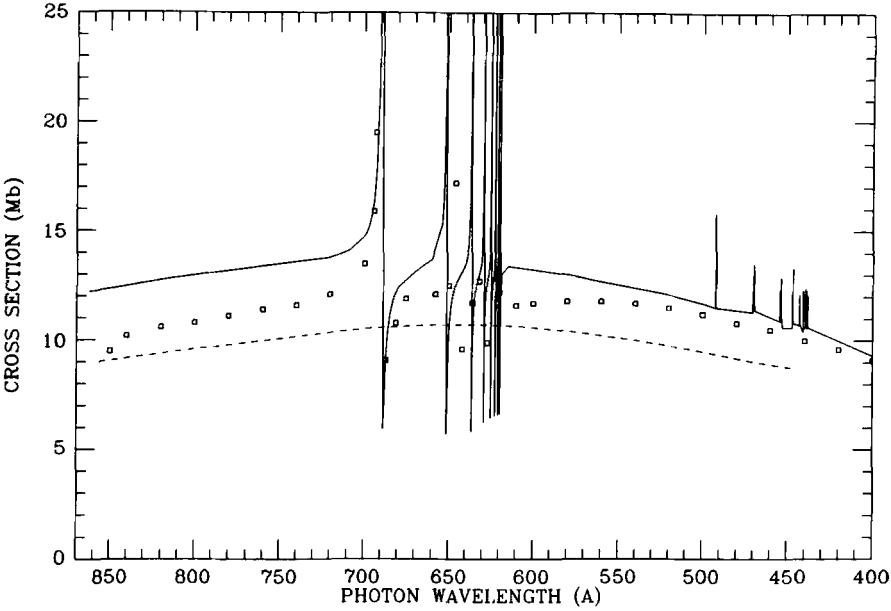

FIG. 2. Photoionisation cross section of nitrogen in the ground state. Theory: —— Bell and Berrington (1991); --- Bates and Seaton (1949). Experiment: □ Samson and Angel (1990).

sections for the production of O^+ in the states $2s^22p^3\,{}^4S°$, $^2D°$, $^2P°$ and $2s2p^4\,{}^4P$ and ^2P states, and the length and velocity formulations of the dipole matrix element were used; as is seen in Figure 1, the length results are in good accord with the more sophisticated theory and experiment at all wavelengths and in excellent agreement for wavelengths shorter than about 500 Å. Indeed these results remain the best of the calculations which do not include resonances; there were many such calculations in the late 1960s and early 1970s, falling into two main groups: those which employed a Hartree–Fock approximation and those which utilised a central potential. The central potential calculations of McGuire (1968) and Starace *et al.* (1974) (Herman–Skillman), Thomas and Helliwell (1970) (Klein–Brueckner), Kahler (1971) and Koppel (1971) (scaled Thomas–Fermi) are compared in Fig. 3 with the experimental data of Angel and Samson (1988). The data of Ganas (1973) (independent particle model) is not shown, but none of this work substantially improved upon that of Dalgarno *et al.* (1964). A similar statement holds true for the Hartree–Fock investigations of Henry (1967) and Starace *et al.* (1974) and the random phase approximation with exchange of Vesnicheva *et al.* (1986) shown in Fig. 4. All of these calculations were based upon a final state representation of $\Psi = A\psi\theta$, where A is an antisymmetrisation operator, ψ is the wave function for the ion and θ is the ejected electron wave function.

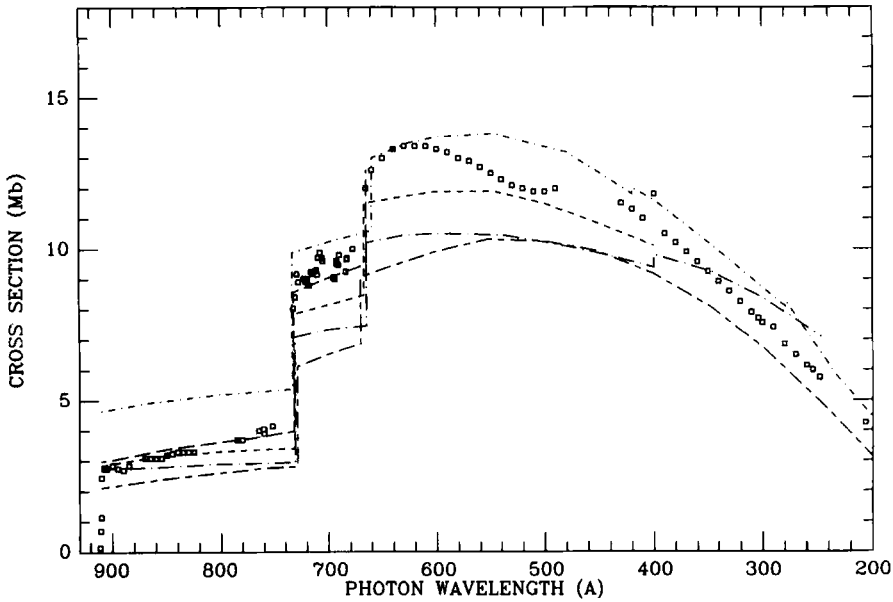

FIG. 3. Photoionisation cross section of oxygen in the ground state. Central field approximation calculations: –– McGuire (1968); —— Thomas and Helliwell (1970); -·- Koppel (1971); — — Kahler (1971) (Kahler data coincides with that of Koppel for $\lambda \leqslant 650$ Å); --- Starace *et al.* (1974). Experiment: □Angel and Samson (1988).

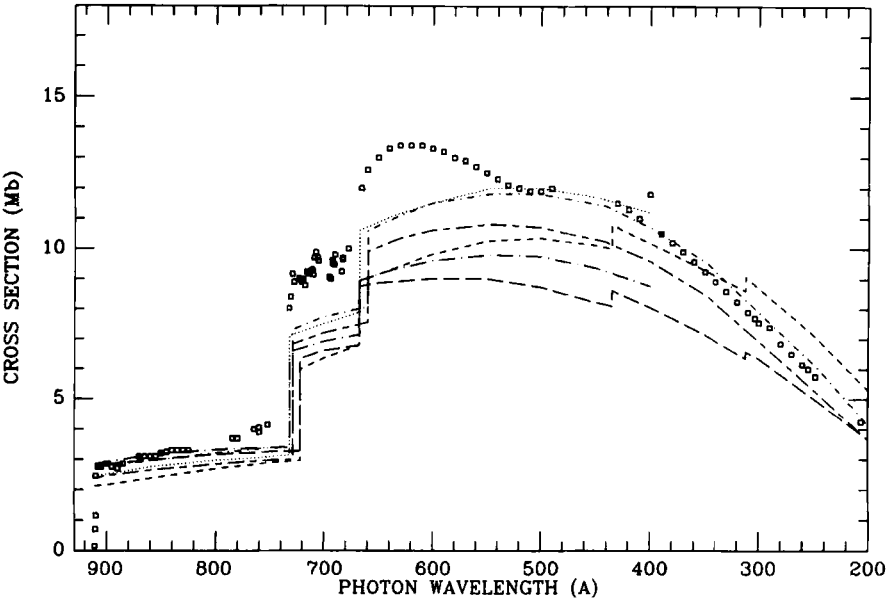

FIG. 4. Photoionisation cross section of oxygen in the ground state. Theory: - - - (length), ——— (velocity), Henry (1967) (HF); ··· (length), – · – (velocity), Starace *et al.* (1974) (HF); - · - (length), – – – (velocity), Vesnicheva *et al.* (1986) (RPAE). Experiment: □ Angel and Samson (1988).

Significant improvement of the theory arose on abandonment of the idea that the final state could be described in terms of an ejected electron moving in the field of just one state of a product ion. The final state representation now takes the form $\Psi = A\,\Sigma_i \psi_i \theta_i$, where the sum is over product ion states i. Not only does this permit greater accuracy but coupling between open and closed channels gives rise to autoionisation resonances. The first calculation in this close-coupling theory including autoionisation was carried out by Henry (1968a). His length formulation results are compared in Figure 5 with the experiment and the more sophisticated R-matrix close-coupling results of Taylor and Burke (1976), Pradhan (1978), and Bell *et al.* (1989), who included seven, seven, and eleven product ion states in the wave function expansion, respectively. Henry's results are some 30% lower than these later values but his resonance positions are in good accord, and he is in best agreement with the most recent experimental data of Angel and Samson (1988)—a point to which we shall return later. As discussed by Bell *et al.* (1989), good agreement exists between theory and experiment in the wavelength region 840–660 Å, which includes the resonances converging on the $^2D^\circ$ and $^2P^\circ$ thresholds. Even more remarkable is the agreement between theory and experiment for the resonance series converging on the ^4P threshold. Figure 6 illustrates the structure found by Angel and Samson (1988), which had not been seen before

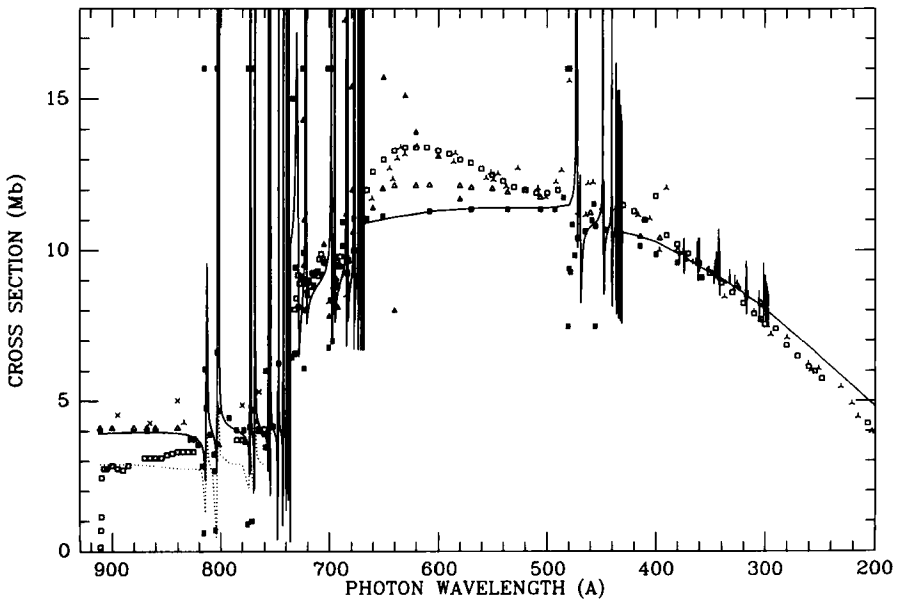

FIG. 5. Photoionisation cross section of oxygen in the ground state. Theory: ··· Henry (1968a); △ Taylor and Burke (1976); ■ Pradhan (1978); — Bell *et al.* (1989). Experiment: x Kohl *et al.* (1978); λ Samson and Pareek (1985); ▲ Hussein *et al.* (1985); □ Angel and Samson (1988).

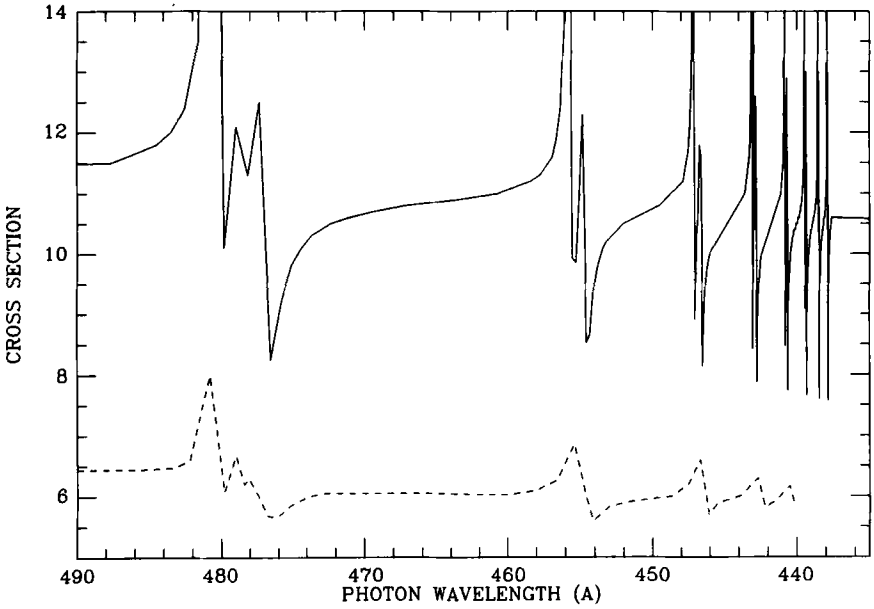

FIG. 6. Photoionisation cross section of oxygen in the ground state; autoionising resonances converging to the ^{4}P threshold. Theory: — Bell *et al.* (1989) Experiment: - - - Angel and Samson (1988).

in photoionisation experiments, and compares it with data from Bell *et al.* (1989) (for which the energy scale has been shifted in order to make the ^{4}P threshold agree with the experimental value). Table I confirms numerically the exceedingly good agreement between theory and experiment for the positions of these autoionising $2s2p^4(^4P)$ np (^{3}D°, ^{3}S°, ^{3}P°) resonances.

We return now to the total cross section and the comparison between theory and experiment presented in Fig. 5. Increased sophistication of the target and free-electron-plus-ion wavefunctions should clearly lead to more accurate theoretical values and as the sophistication increases some convergence of the theoretical results should appear. This is indeed the case—comparison of the values obtained by Taylor and Burke (1976), Pradhan (1978) and Bell *et al.* (1989) not only show convergence but suggest that theoretically the total cross section is now known to better than a few percent at all wavelengths considered. However, comparison with experiment reveals two major discrepancies neither of which has yet been resolved. Above the ^{4}S° threshold, the latest experimental data (Angel and Samson, 1988) and the earlier values of Comes *et al.* (1968) (not shown in Fig. 5 but that agree closely with the results of Henry (1968a)) lie considerably lower than the *R*-matrix results (by almost 50% at threshold) and the shape of the cross section with

TABLE I
AUTOIONISING $2s2p^4(^4P)$ np ($^3D°$, $^3S°$, $^3P°$) RESONANCE SERIES BELOW THE ^4P THRESHOLD IN ATOMIC OXYGEN

Resonance State	Wavelength (Å)			
	(1)	(2)	(3)	(4)
$3p\,^3D°$	480.8	480.7	480.6	479.3
$3p\,^3S°$	479.0	478.8	478.6	477.6
$3p\,^3P°$	478.1	477.1	476.1	475.6
$4p\,^3D°$	455.4	455.7	455.6	455.2
$4p\,^3S°$		454.6	455.3	454.8
$4p\,^3P°$		454.5	454.7	454.0
$5p\,^3D°$	446.7	447.0		446.8
$5p\,^3S°$		446.7		446.6
$5p\,^3P°$		446.5		446.3
$6p\,^3D°$	442.7	442.9		
$7p\,^3D°$	440.5	440.7		

Columns: (1) Angel and Samson (1988); (2) Bell *et al.* (1989); (3) Pradhan (1978); (4) Taylor and Burke (1976).

wavelength is also at variance with theory. The second region of discrepancy lies above the $^2P°$ threshold; Angel and Samson (1988) find a peak in the cross section and Hussein *et al.* (1985) also find values much larger than theory (with the exception of their very small value at 640 Å). Bell *et al.* (1988) suggested that the latter discrepancy may be due to difficulty in eliminating molecular oxygen contamination in the beam or in the work of Angel and Samson (1988) excited state oxygen contamination.

In conclusion, theory suggests that the cross section for photoionisation of ground state oxygen is known to better than 5%. Further experimental work is desirable for several reasons: (1) to resolve the $^4S°$ threshold discrepancy; (2) to resolve the preceding $^2P°$ threshold discrepancy; (3) to provide experimental results for the widths of resonances.

B. EXCITED STATES

Little theoretical and experimental data exists for photoionisation of excited states of oxygen in comparison with the ground state case. No experimental data seem to exist for photoionisation of the excited metastable $2s^22p^4\ ^1D$ and ^1S states, and until recently the theoretical work was relatively unsophisticated. Henry (1967) employed a close-coupling approximation but

included only open channels in his wavefunction expansion. Thomas and Helliwell (1970) used a central potential model based upon the Klein–Brueckner potential and Koppel (1971) adopted the scaled Thomas–Fermi potential method. Wide divergence exists for both ^{1}D and ^{1}S states both with regard to magnitude and shape of cross section with variation in photon energy, and significantly none of these calculations permitted autoionisation. Inclusion of coupling between closed and open channels by Bell *et al.* (1989) in an 11-state *R*-matrix calculation revealed that, for both states, the cross section is dominated by a $2s2p^5\ ^1P^\circ$ Coster–Kronig resonance (see Fig. 7). Further work on these states is desirable since it is not possible from the present knowledge to give an accuracy to the currently most accurate data (that of Bell *et al.*, 1989).

Photoionisation of the higher excited states $2p^33s\ ^{3,5}S^\circ$, $2p^33p\ ^{3,5}P$ have been treated using an 11-state *R*-matrix method by Bell *et al.* (1990). For both the $3s\ ^3S^\circ$ and $3s\ ^5S^\circ$ it was found that the cross section remained relatively small until the $3s\ ^2P$ and $3s\ ^4P$ thresholds, when the cross sections suddenly increased, passed through a maximum and then decreased as the photon energy increased. Analysis reveals that the large cross section results from a transition in which the 2p electron is ejected into the continuum with the 3s electron behaving like a "spectator" electron.

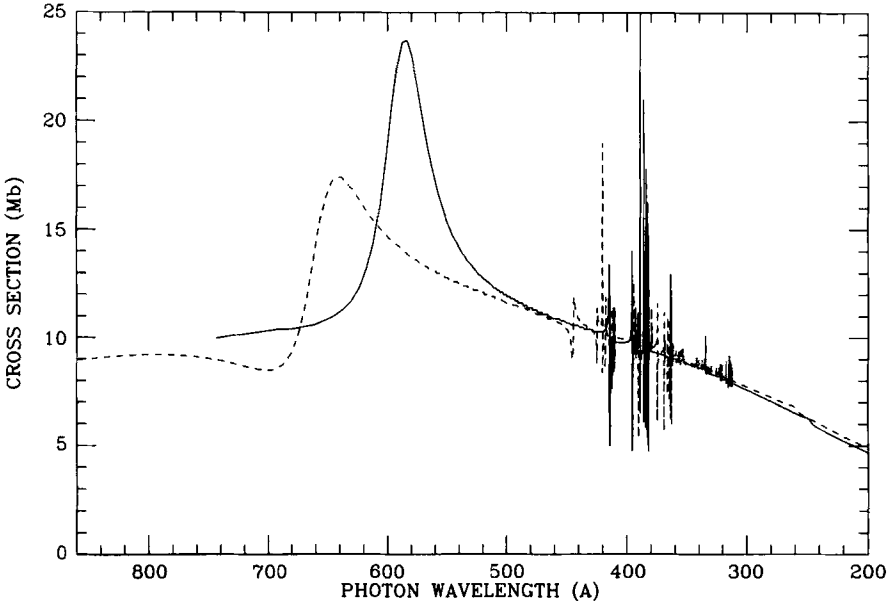

FIG. 7. Photoionisation cross section of oxygen in the $2s^22p^4\ ^1$D and ^{1}S states. Theory: Bell *et al.* (1989) —, ^{1}D; - - -, ^{1}S.

The $3p\,^{3,5}P$ states both reveal a minimum, and we shall concentrate on the $3p\,^3P$ state for which the minimum was first found by Saxon *et al.* (1989). Their calculation was based on a variational R-matrix approach for electron–molecule scattering and was restricted in that configurations describing $e^- + O^+$ (^2P, ^2D) were not included in their configuration-interaction expansion so that autoionising resonances were automatically excluded. Figure 8 compares the results of Saxon *et al.* (1989) and Bell *et al.* (1990). Also shown is the value obtained by Dixit *et al.* (1988) as part of a study of the two-photon-resonant–three-photon ionisation of atomic oxygen using quantum defect theory to calculate the relevant atomic parameter and an experimental data point (Bamford *et al.*, 1986). For energy values greater than about 0.12 Ry above the threshold, the Saxon *et al.* data falls below that of Bell *et al.* but the more serious discrepancy exists as one approaches the threshold where the Saxon *et al.* value is about 60% greater than that of Bell *et al.*

This discrepancy may be investigated by examining the individual $^3S°$ and $^3D°$ partial cross sections that contribute to the total at these low energies and making use of a continuity relationship between bound–bound and bound–free absorption. Seaton (1978) and Dubau and Seaton (1984) have

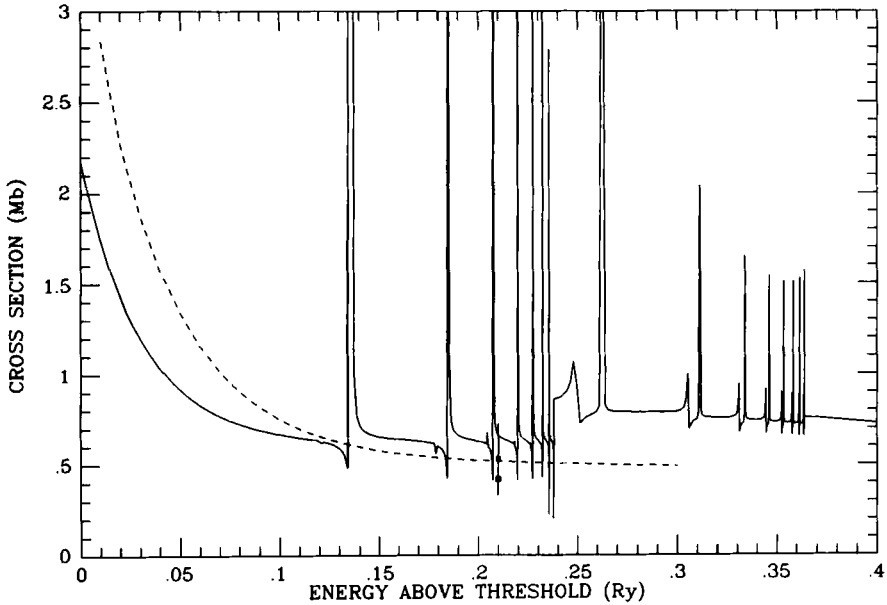

FIG. 8. Photoionisation cross section of oxygen in the $3p\,^3P$ state: — Bell *et al.* (1990); --- Saxon *et al.* (1989); □ Dixit *et al.* (1988); ■ Bamford *et al.* (1986).

shown that the differential oscillator strength per Rydberg, $df/d\varepsilon$, derived by considering bound–bound absorption and given by

$$\frac{df}{d\varepsilon} = \frac{(n-\mu)^3}{2} f(b \rightarrow a)$$

joins smoothly to the differential oscillator strength derived from the photoionisation cross section and given by

$$\frac{df}{d\varepsilon} = \frac{1}{4\pi^2 \alpha a_0^2} \sigma$$

where f is the oscillator strength for the bound–bound transition from state b to state a, n and μ are the principal quantum number and quantum defect, respectively, for state a, σ is the cross section and α is the fine structure constant. In the present case, state b corresponds to $2p^3(^4S^\circ)3p\ ^3P$, and we consider as final states a $2p^3(^4S^\circ)ns\ ^3S^\circ$ and $2p^3(^4S^\circ)nd\ ^3D^\circ$. The term $df/d\varepsilon$ evaluated from the preceding two expressions is presented in Fig. 9; very accurate bound–bound oscillator strengths from Bell and Hibbert (1990) and Butler and Zeippen (1991) have been employed in the calculation of $df/d\varepsilon$ below the threshold and the photoionisation cross sections of Bell *et al.* (1990) and Saxon *et al.* (1989) above the threshold. The minimum in the $^3D^\circ$ final symmetry is apparent, and for this symmetry the largest difference between the results of Bell *et al.* (1990) and Saxon *et al.* (1989) occurs. The "below threshold" $df/d\varepsilon$ clearly indicates better continuity with the work of Bell *et al.* (1990) and would suggest that these results are more accurate than those of Saxon *et al.* (1989).

III. Photoionisation of Atomic Nitrogen

A. Ground State

As one might expect the history of theoretical calculations of the photoionisation cross section for ground state nitrogen closely follows that for oxygen. However, after the early work of Bates and Seaton (1949) (see Fig. 2) it took almost 30 years before a calculation was performed that included autoionisation. Dalgarno and Parkinson (1960) employed a Hartree bound state and hydrogenic continuum functions to calculate the cross section, modified the treatment of Bates (1946) to include the dipole velocity formulation and also took into account absorption due to inner shell electrons. Henry (1966, 1968b) used the Hartree–Fock approximation and

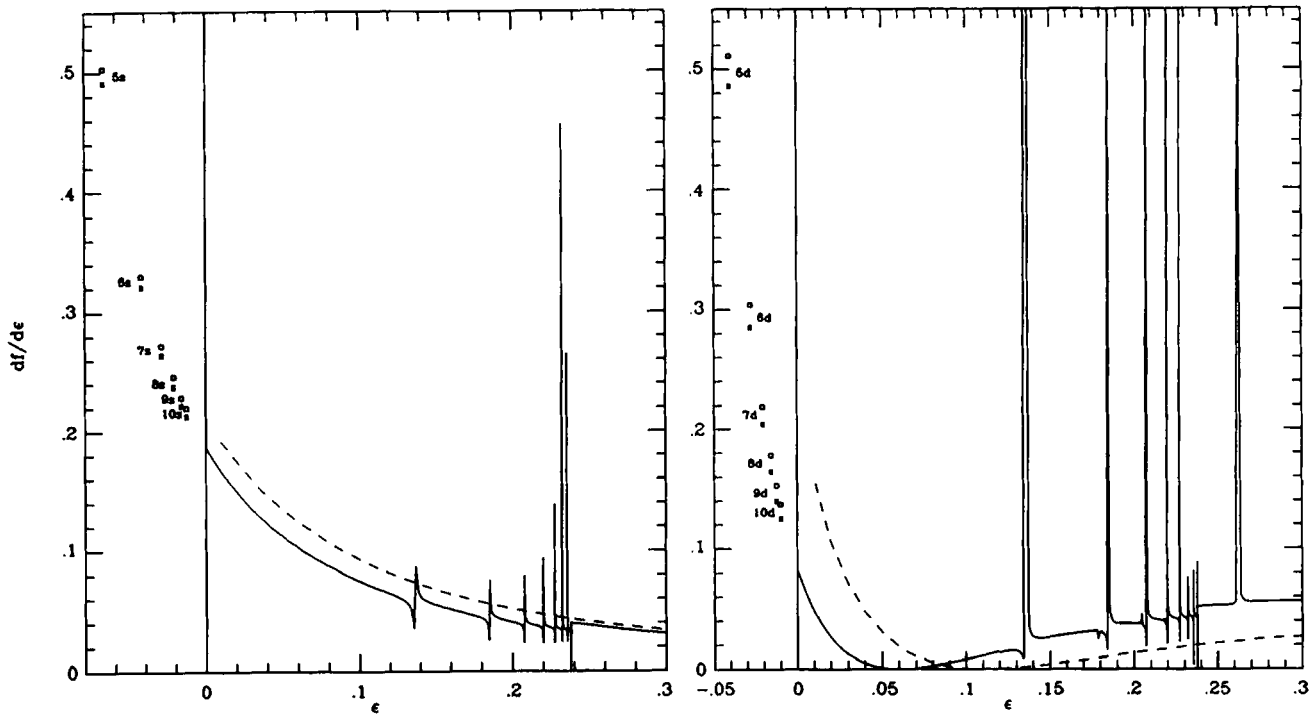

FIG. 9. Differential oscillator strength for the transitions in oxygen $3p\,^3P-{}^3S^\circ$ (lefthand figure) and $3p\,^3P-{}^3D^\circ$ (righthand figure). The energy above the threshold, ε, is in Rydbergs: — Bell *et al.* (1990); - - - Saxon *et al.* (1989); □, ■ derived from oscillator strengths taken from Bell and Hibbert (1990) and Butler and Zeippen (1991), respectively.

included a single-state close-coupling representation of the final continuum and so did not take into account resonances. A number of model potential calculations then followed: McGuire (1968) (Herman–Skillman), Thomas and Helliwell (1970) (Klein–Brueckner), Kahler (1971) and Koppel (1971) (scaled Thomas–Fermi), and Ganas (1973) (independent particle). These calculations are presented in Figs. 10 and 11, where they are compared with the currently most sophisticated results (Bell and Berrington, 1991—note that the resonance structure has been averaged out). Kahler's data is not given since it almost coincides with that of Koppel (1971). Considerable disagreement—both quantitative and qualitative—is apparent with only the data of Koppel (1971) in accord with the most recent work (Bell and Berrington, 1991). Figure 11 also contains the results of Cherepkov *et al.* (1974), who employed a random phase approximation with exchange but neglected the strong final-state coupling between the 2p and 2s photoejections and failed to predict the $2s2p^3\ ^5S^\circ np^4P$ resonances. These resonances were first considered theoretically by Hempe (1978) using a many-channel quantum defect method and later by Le Dourneuf *et al.* (1979) and Bell and Berrington (1991), who both employed the *R*-matrix method. Figure 12 presents data from these three calculations and Table II gives a

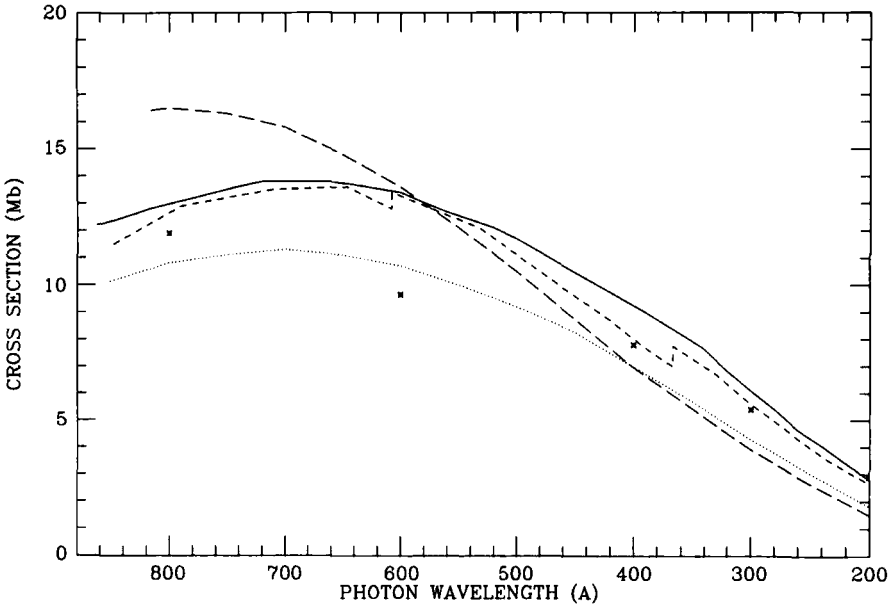

FIG. 10. Photoionisation cross section of nitrogen in the ground state. Central-field approximations: x McGuire (1968); ··· Thomas and Helliwell (1970); - - - Koppel (1971); – – Ganas (1973). *R*-matrix: — Bell and Berrington (1991).

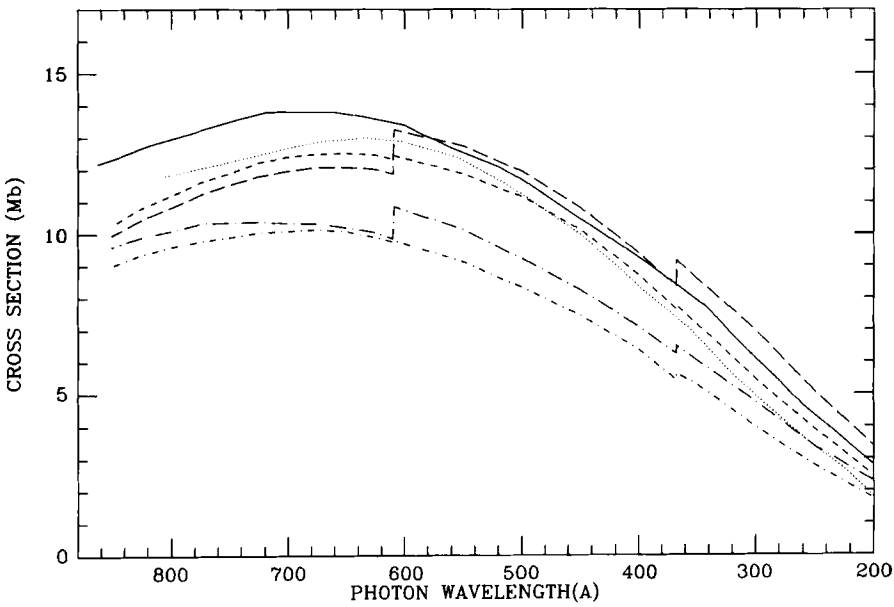

FIG. 11. Photoionisation cross section of nitrogen in the ground state: - - (length, - · - (velocity) Henry (1966); – – (length), – · – (velocity) Henry (1968b); · Cherepkov *et al.* (1974); — Bell and Berrington (1991).

TABLE II

THE $N^{**}(2s2p^3\ ^5S^\circ\ np\ ^4P)$ RESONANCE PROFILES

	$n = 3$				$n = 4$			
	(1)†	(2)	(3)	(4)	(1)	(2)	(3)	(4)
E_n(Ryd)	1.310	1.323	1.322	1.316	1.405	1.410	1.399	1.408
$\Gamma(10^{-3}$ Ryd)	1.51	2.03	1.65	2.02 ± 0.22	0.67	0.70	0.79	0.73 ± 0.22
q		-1.70 $(-1.95)^{\#}$	-2.03	-1.7 ± 0.1		-1.85 (-2.09)	-1.91	-1.7 ± 0.1
r^2		0.59 (0.60)	0.52	0.51 ± 0.04		0.61 (0.62)	0.56	0.58 ± 0.04

†(1) Hempe (1978); (2) Le Dourneuf *et al.* (1979); (3) Bell and Berrington (1991); (4) Dehmer *et al.* (1974).

Values obtained for q and r^2 using the velocity formulation are given in parentheses.

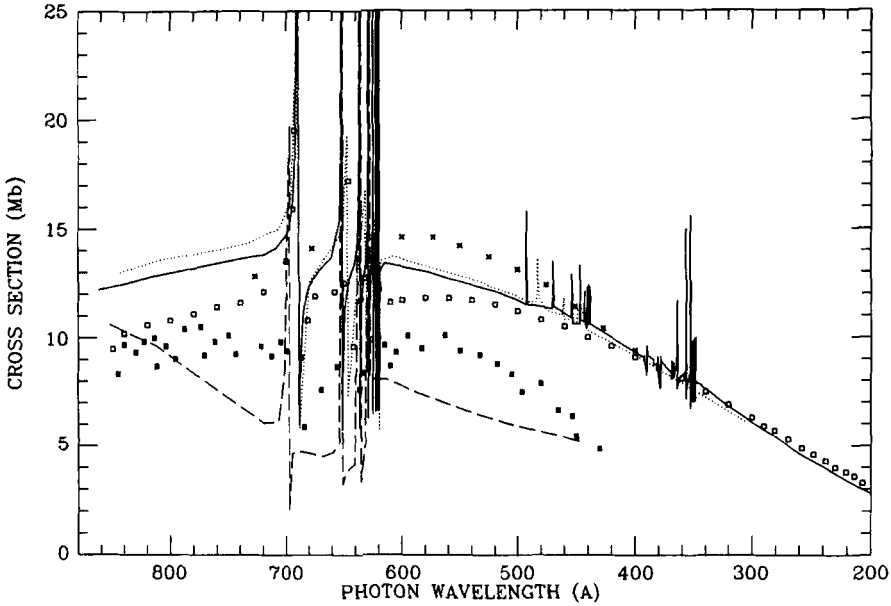

FIG. 12. Photoionisation cross section of nitrogen in the ground state. Theory: — Hempe (1978); ·· Le Dourneuf *et al.* (1979);—Bell and Berrington (1991). Experiment ⌘ Ehler and Weissler (1955); ■ Comes and Elzer (1967, 1968); □ Samson and Angel (1990).

comparison of the $2s2p^3\ ^5S°np\ ^4P$ resonance profiles. In Table II, the resonance position E_n, width Γ, line profile index q and correlation coefficient ρ^2 (see Burke and Taylor, 1975, for a definition of these parameters) are compared with the experimental results of Dehmer *et al.* (1974), who studied the series using a continuum light source. The energy positions used by Dehmer *et al.* (1974) had been determined earlier by Carroll *et al.* (1966), using conventional photographic absorption techniques. The agreement between theory and experiment is satisfactory, noting that Dehmer *et al.* set q to the value of -1.7 to parameterize their $n = 4$ data.

The total cross section (Fig. 12) obtained by Hempe (1978) differs in both shape and magnitude from the results of Le Dourneuf *et al.* (1979) and Bell and Berrington (1991). The good agreement between the two R-matrix calculations—five state of Le Dourneuf *et al.* and eight state of Bell and Berrington—suggests that this method has "converged" and that theoretically the cross section is known to an accuracy of a few percent. Such a statement is also supported by the recent experimental data of Samson and Angel (1990) for wavelengths shorter than 520 Å. At longer wavelengths discrepancy between theory and experiment is apparent. However, Samson and Angel (1990) normalised their data in the wavelength region 520–852 Å

by employing the Thomas–Reiche–Kuhn sum rule for the total oscillator strength. Bell and Berrington (1991) have pointed out that more accurate discrete oscillator strengths give a contribution to this sum of 0.58 in comparison with the value of 0.96 used by Samson and Angel. Therefore the experimental data should be renormalised upward and so remove the discrepancy between the *R*-matrix results and experiment.

B. Excited States

In contrast to oxygen, sophisticated theoretical data was available for the metastable states of nitrogen in 1980. The early central potential calculations of Thomas and Helliwell (1970) and Koppel (1971) were superseded by the five-state *R*-matrix work of Zeippen *et al.* (1980). Figure 13 shows that the data of Koppel agree closely with that of Zeippen *et al.* and the more sophisticated 13-state *R*-matrix calculation of Bell *et al.* (1992) but of course is incapable of obtaining the $2s2p^4\ ^2P$ resonance that dominates the cross section for both the $2s^22p^3\ ^2D^\circ$ and $^2P^\circ$ states. The close accord between the two *R*-matrix calculations suggests that for both states the cross section is known to an accuracy of a few percent.

Bell *et al.* (1992) have also obtained data for the $2s^22p^33s\ ^{4,2}P$, $2s2p^4\ ^4P$ and $2s^22p^23p\ ^2S^\circ$ excited states. Again the effect of increase in the cross section due to photoejection of the 2p electron with the 3s electron behaving as a

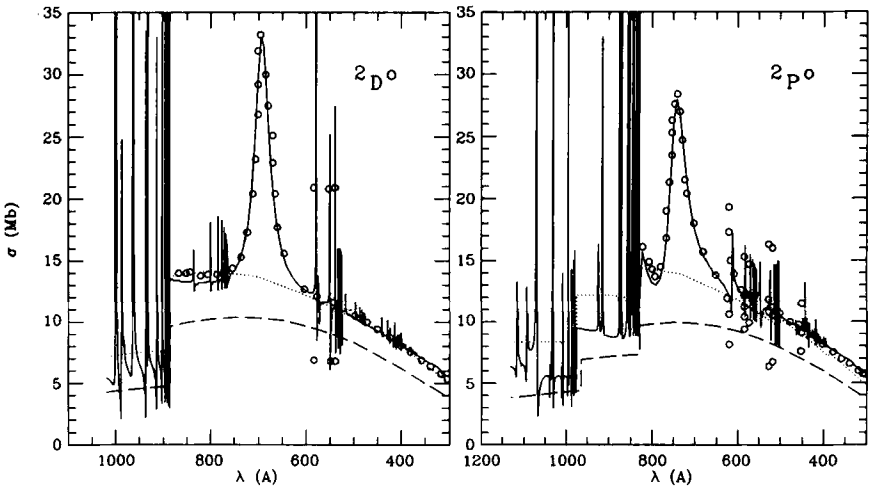

FIG. 13. Photoionisation cross section of nitrogen in the $2s^22p^3\ ^2D^\circ$ and $^2P^\circ$ states: —— Thomas and Helliwell (1970); ··· Koppel (1971); ○ Zeippen *et al.* (1980) —— Bell *et al.* (1992).

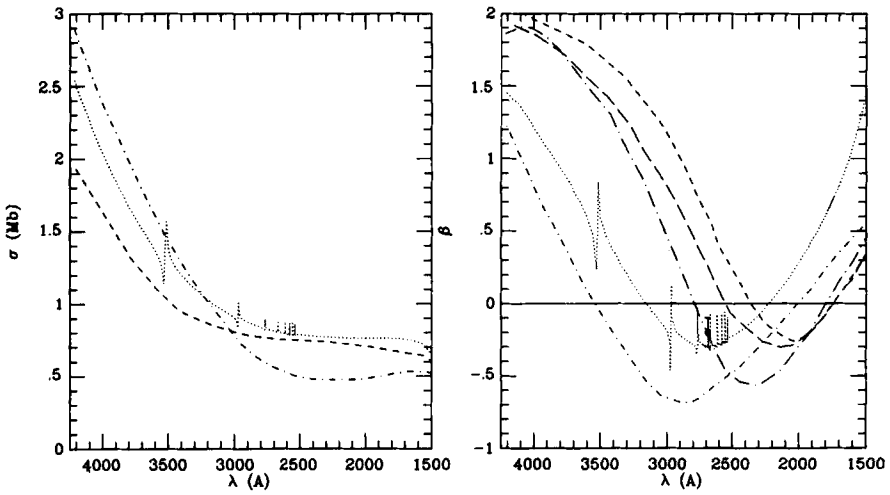

FIG. 14. Photoionisation cross section and photoelectron angular distribution asymmetry parameter β for the $3p\,^2S^\circ$ excited state of nitrogen. Cross section: ··· Bell *et al.* (1992); -·- Theodosiou (1988); --- Manson (1988). Asymmetry parameter: ··· Bell *et al.* (1992); --- (length), —— (velocity), Nahar and Manson (1989); -·- (HS ground state), –·– (HS excited state) Theodosiou (1988). Experiment: ● Pratt *et al.* (1987).

"spectator" electron is found. Of these states, comparison with other data may be made for the $3p\,^2S^\circ$ state, and this state is also of interest because it is the only excited state of either oxygen or nitrogen for which an experimental investigation has been performed for the asymmetry parameter, β. Figure 14 compares the low-energy total cross-section data of Bell *et al.* with the central-field Hartree–Slater calculations of Manson (1988) (using a ground-state potential) and Theodosiou (1988) (using the excited-state potential). Clearly the central-field results are sensitive to the potential used, but all three calculations confirm the existence of a minimum. This sensitivity is more marked in the case of the β-parameter, where, as is seen in Fig. 14, wide divergence of results exists. The single experimental point of Pratt *et al.* (1987) is unfortunately insufficient to discriminate between the Hartree–Slater, *R*-matrix and Hartree–Fock results (Nahar and Manson, 1989).

IV. Conclusion

In this short chapter we have concentrated mainly on total cross sections. It is recognised that other theoretical data is available. The opacity project has produced total cross sections for both ground and excited states, and data are

available for partial cross sections as well as angular distributions of photoelectrons. It is our opinion that, for both elements, the theoretical data are of sufficient accuracy for most applications. Further experimental work is clearly required particularly with a view to resolving the difficulties discussed for ground-state oxygen and for the $3p\,^2S^\circ$ state of nitrogen.

In the conclusion to his paper of 1946, David Bates made several pertinent comments—we abstract but two: "The main future development of the theory probably lies in attempting to achieve increased accuracy" and "The great computational labour involved is likely severely to limit any programme." This review chapter clearly reflects the first statement, and without large computers (and a vast amount of computer time) the second statement would also have remained true.

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