Measurement of the photodetachment cross section of the negative ion of fluorine

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The photodetachment cross section of F^- was obtained experimentally from a stationary arc plasma of SF_6 at atmospheric pressure. The temperature was determined by spectroscopy. The electron number density was measured by two-wavelength laser interferometry and by emission spectroscopy. Departures from local thermodynamic equilibrium were taken into account. The variations of the photodetachment cross section of F^- versus wavelength were obtained by measuring the absolute intensity of the continuum radiation emitted by the plasma and are compared with results from the literature.

I. INTRODUCTION

The photodetachment cross section σ_{F^-} of the negative ion F^- has been determined in a few experimental studies and in numerous theoretical studies which do not all agree perfectly. The values of σ_{F^-} generally used are those which Mandl[1] obtained by absorption spectroscopy of a plasma of CsF vapors in a shock tube. They are used as a standard for the comparison of the theoretical values proposed in the literature. Using emission spectroscopy, Popp[2] studied the affinity continuum of F^- ions of an arc plasma in SF_6. The values of σ_{F^-} he found are lower than those of Mandl. Popp's experimental method led him to underestimate the intensity of the affinity continuum. The only other experimental results available are those of Berry and Reimann;[3] however, they are of little use since they concern the region near the threshold.

The theoretical studies of Moskvin[4] gave very high values probably due to the simplification hypotheses used: the states of continuum are represented by plane waves. Although it is semiempiric, Robinson and Geltman's model[5] is more sophisticated. The free and bound states are calculated in a central field in which an adjustable parameter allows the binding energy of the negative ion to be taken into account. The values obtained are a little higher than those of Mandl:[1] They can be superimposed after multiplication of Robinson and Geltman's data by a factor of 0.7. Using the many-body perturbation theory, Ishihara and Foster[6] found values of σ_{F^-} in good agreement with those of Mandl. Rescigno *et al.*[7] calculated σ_{F^-} using the method of Stieltjes imaging from a finite number of transition energies and oscillator strengths obtained in a calculation employing discrete basis functions only. The values are a little higher than those of Ishihara and Foster or Mandl. The disagreement is most noticeable at low energies. Rescigno *et al.*[7] consider the agreement to be acceptable but regret the lack of experimental data. More recently Clodius *et al.*[8] calculated the photodetachment cross sections of 20 elements by the zero-core-contribution model taking into account the levels of the fine structure and the highest electron energy states of the anions and neutral atoms. The results obtained are, in general, in good agreement with experimental data. For fluorine, however, the values of σ_{F^-} are higher than those proposed by Mandl[1] and the shape of the curve is not quite the same. The only experimental results which allow theoretical data to be evaluated are those of Mandl. Recent studies carried out on an arc discharge in SF_6 at atmospheric pressure[9,10] in association with recent calculations of Biberman's coefficient of the S^+ ion[11] allow the accurate determination of σ_{F^-}. This is the purpose of the present work.

II. PRINCIPLE OF σ_{F^-} DETERMINATION

The radiation spectrum emitted by the positive column of an arc in SF_6 at atmospheric pressure is given in Fig. 1. This spectrum was obtained with a low-resolution spectroscope along an axis perpendicular to the discharge axis. The axial temperature was around 10 000 K. No corrections have been made to take into consideration the wavelength response of each element of the optical arrangement.

The continuum spectrum is the sum of several components:

the recombination continuum of the positive ions which spreads across the whole spectral range studied,

the bremsstrahlung (free-free radiation) which makes a

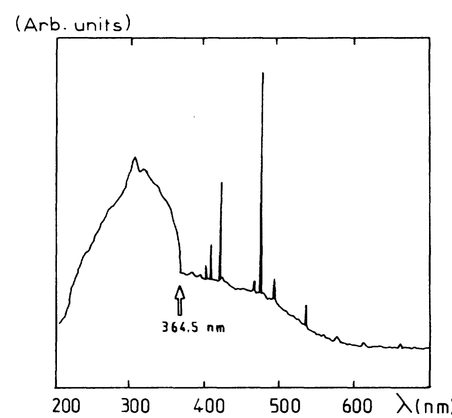

FIG. 1. Continuum emitted by a SF_6 arc plasma (axial temperature of the discharge, 10 000 K; atmospheric pressure).

significant contribution to the infrared part of the spectrum, and

the negative ion attachment continuum which appears at a given threshold value and spreads to the ultraviolet part of the spectrum.

The principle of the electron attachment reaction is

$$A+e^{-}\rightleftarrows A^{-}+h\nu \ .$$

The emission coefficient ε_{att}, defined as the radiant energy emitted by a unit volume per unit of time, per unit of solid angle, and per unit of wavelength, can be expressed as a function of the wavelength λ and the temperature T by the expression[12]

$$\begin{aligned}\varepsilon_{att}&=B(\lambda,T)\sigma_{A^-}N_{A^-}\left[1-\exp\left[-\frac{hc}{\lambda kT}\right]\right]\\&=2\frac{hc^2}{\lambda^5}\exp\left[-\frac{hc}{\lambda kT}\right]\sigma_{A^-}(\lambda)N_{A^-}\ ,\end{aligned} \quad (1)$$

where $B(\lambda,T)$ is Planck's function, $\sigma_{A^-}(\lambda)$ the photodetachment cross section of the ion A^-, N_{A^-} the number density of ion A^-, c the velocity of light, h Planck's constant, and k Boltzmann's constant.

To determine σ_{A^-} it is necessary to know the temperature T of the medium; know the number density of the negative ions N_{A^-}; and be able to isolate, from the spectrum shown in Fig. 1, the contribution of the attachment continuum ε_{att}. It is therefore these three parameters $\varepsilon_{att}(\lambda,T)$, T, and N_{A^-} which we shall determine. T can be obtained directly from the spectroscopic measurements. N_{A^-} can be obtained from the values of T and of the electron number density N_e by using Saha's law. The ε_{F^-} threshold appears at 365 nm. Note that fine analysis of the spectrum in Fig. 1 reveals a second threshold at 359.5 nm arising from the ground-state configuration of $F(^2P_{3/2,1/2})$. In the wavelength range under consideration (between 200 and 365 nm), the continuum is the sum of ε_{F^-},ε_{S^-} (which is due to the formation of S^- ions), ε_{S^+} and ε_{F^+} positive ion recombination continua, and ε_{brems} arising from bremsstrahlung. We shall analyze each of these terms. It can be noted that in Fig. 1 there is a wide structure at about 300 nm which could correspond to a shape resonance level of the F^- ion. This emission will not be taken into account in the measurement of the F^- affinity continuum.

III. EXPERIMENTAL SETUP

The experimental setup has been described elsewhere[9,10] and we shall only mention the essential points. The arc chamber is of "cascade" type. It is constituted of eight water-cooled hollow disks; the electrical insulation is provided by alumina rings. The central hole, which determines the discharge channel, is of diameter 5 mm. The tungsten electrodes, with a 7-cm gap, are eccentric to allow end-on measurements and are protected by a flow of argon. Side-on measurements (requiring Abel inversion to obtain local values) were made at an equal distance from both electrodes. Suitably placed windows prevent argon from penetrating the measurement zone and the SF_6 from reaching the electrodes. The arc current is provided by a series of three regulated power supplies delivering up to 100 A under 350 V.

The measurements were carried out by spectroscopy and by laser interferometry at two wavelengths. The experimental arrangement has been described in detail by Kafrouni.[13] The method used has been detailed in Ref. 10. Let us just mention that the measurements of electron density by laser interferometry were carried out end-on without argon at the electrodes. It was possible to dispense with this protection because the measurement time was very short (a few seconds).

For the measurement of absolute radiation intensities a tungsten ribbon lamp was used for calibration for $\lambda > 310$ nm and a standard graphite electrode arc for $\lambda < 340$ nm. The graphite electrodes were of the type NORIS H for the 6-mm-diam anode and type NORIS D for the 7-mm-diam cathode. The spectral emission of these graphites has been widely studied.[14] The total radiation emitted by the standard arc and measured end-on is the sum of the radiation from both the anode crater and that due to the arc plasma. Owing to the stability and the symmetry of this arc, the radiation due to the anode crater only can be deduced by subtracting the contribution of the plasma (measured side-on) from the total radiation. We performed this experiment and our results are in very good agreement with those of Magdeburg and Schey.[14] The contribution of the plasma represents 2% of the overall signal at $\lambda=300$ nm and about 18% at $\lambda=250$ nm. So a correction was made to take into account the plasma emission near the anode, the reflectivity of the mirrors, and the transmission coefficient of the lenses. The signals from the two standards link up well giving superimposable curves between 310 and 340 nm.

All the experiments were carried out at two arc currents, 25 and 30 A. As the current increases, the signals become more intense and the conditions for thermodynamic equilibrium and spatial stability of the discharge are better fulfilled, but the intensity of the affinity continuum is low compared with that of recombination. This is unfavorable to the accuracy of the measurements.

IV. DETERMINATION OF TEMPERATURE T

The assumptions made for the measurement of T have been justified in discussions on local thermodynamic equilibrium (LTE) developed elsewhere.[15] Two methods were used: the Boltzmann's diagram method, using 11 lines of ionized sulfur, and a method proposed by Schulz-Gulde[16] based on the measurement of the ratio of the absolute intensity of two lines: the 545.4-nm line of ionized sulfur ($4s\ ^4P$—$4p\ ^4D°$ transition) and the 469.5-nm triplet of neutral sulfur ($4s\ ^3S°$—$5p\ ^3P$ transition). The results are presented in Fig. 2.

V. DETERMINATION OF THE INTENSITY OF THE RECOMBINATION CONTINUUM ε_{rec}

The principle of the emission process of the recombination continuum is, for radiative recombination,

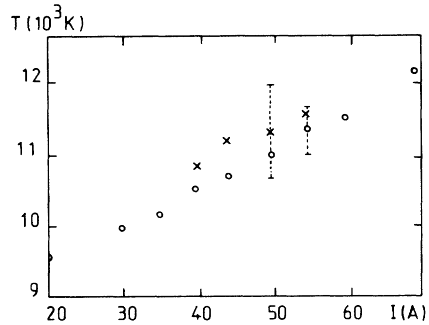

FIG. 2. The temperature along the discharge axis vs the current intensity, obtained from the Boltzmann diagram (×) and by two-line $\mathscr{I}$(S II)/$\mathscr{I}$(S I) method (○).

$$A^+ + e^- \leftrightarrows A + h\nu \ .$$

The coefficient of emission can be expressed in terms of the wavelength λ and temperature T as[12]

$$\varepsilon_{rec}(\lambda,T) = C_1 \frac{g}{Q_1(T)} \frac{N_e N^+}{\lambda^2 T^{1/2}} \times \xi(\lambda,T) \left[1 - \exp\left[\frac{-hc}{\lambda k T} \right] \right] , \quad (2)$$

where $C_1 = 16\pi(e^2/4\pi\varepsilon_0)^3/3c^2(6\pi m^3 k)^{1/2} = 1.632 \times 10^{-43}$ (in SI units), g is the statistical weight of the fundamental level of the ion, $Q_1(T)$ the partition function of the ion, N_e the electron number density, N^+ the number density of positive ions, and $\xi(\lambda,T)$ a quantum-mechanical correction factor introduced by Biberman and Norman[17] in the calculation of the photoionization cross section of non-hydrogen-like atoms.

The problem is therefore to determine ε_{rec} over the spectral range 200 to 365 nm. To do this the following must be known: the nature of the positive ions in the discharge and their number density, the value of the coefficient ξ, and the number density of the electrons.

A. The nature and density of the positive ions in the discharge

The ionization potential of the sulfur atom is 10.358 eV. That of the fluorine atom is 17.420 eV. For temperatures below 12 000 K the positive ions will therefore be mainly S^+. Thus

$$\frac{N_{F^+}}{N_{S^+}} = 7.6 \times 10^{-2}, \ \text{for } T = 12\,000 \text{ K}$$

$$\frac{N_{F^+}}{N_{S^+}} = 2.3 \times 10^{-2}, \ \text{for } T = 11\,000 \text{ K} \ .$$

The error brought about by assuming that $N_e = N_{S^+}$ is, for $T = 11\,000$ K, lower than 2.3%. Indeed

$$\varepsilon_{rec}(\text{total}) = \varepsilon_{rec}(S^+) + \varepsilon_{rec}(F^+) \ ,$$

$$\varepsilon_{rec}(\text{total}) = f(\lambda,T,N_e) \left[\frac{g(F^+)}{Q(F^+)} \xi(F^+) N_{F^+} + \frac{g(S^+)}{Q(S^+)} \xi(S^+) N_{S^+} \right] ,$$

where $N_e = N_{F^+} + N_{S^+}$.

If we consider that $N_e = N_{S^+}$, we only introduce an error if $(g/Q)\xi$ is different for F^+ and for S^+. Now, for the values of λ studied, Biberman and Norman's factor ξ is close to 1 and the ratio g/Q for F^+ is near that for S^+. The error introduced to ε_{rec} is consequently lower than 1% and that introduced to N_e lower than 0.5%. We shall therefore consider that $N_e = N_{S^+}$, which also gives us a figure for the density of S^+ ions present in the plasma.

B. Biberman and Norman's factor

The theoretical values of Biberman and Norman's factor determined by Hofsaess[11] have been cross-checked experimentally.[9] The agreement was good in the part where checking was possible ($\lambda > 365$ nm). We therefore used the values presented in Fig. 3. Below 250 nm the relative inaccuracy of ξ is compensated by the low intensity of the recombination continuum which is less than that of the affinity continuum. The calculations of the recombination continuum are made over the range $200 < \lambda < 500$ nm.

C. Electron number density

The electron density along the discharge axis has been determined using three methods:[10] with two-wavelength laser interferometry, from the S^+ ion recombination continuum, and by measuring the broadening of the sulfur lines by the Stark effect.

The results are presented in Fig. 4 as a function of the arc current I. It can be seen that as I (and T) rises the difference between the experimental value of n_e and its

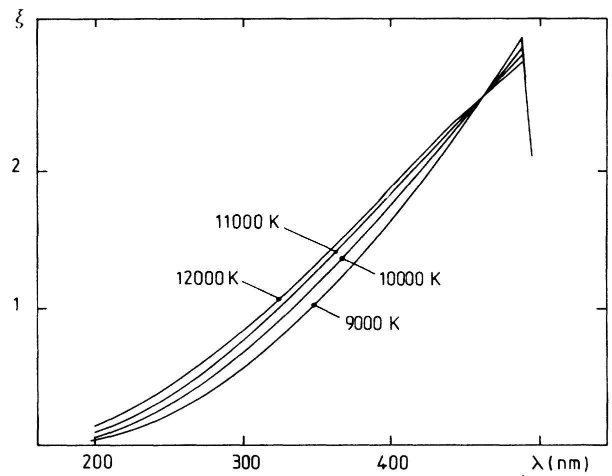

FIG. 3. The Biberman and Norman factor vs the wavelength for four values of T (Ref. 11).

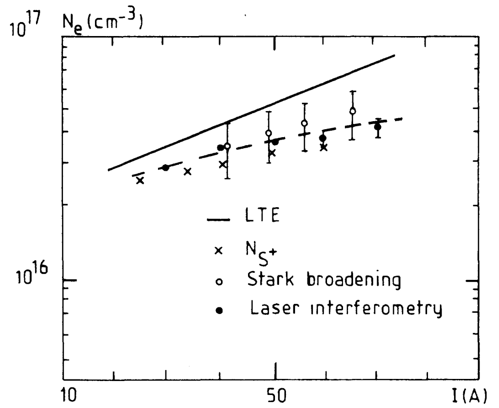

FIG. 4. The electron number density N_e along the discharge axis vs the current intensity.

theoretical value at LTE increases. This is mainly due to the phenomenon of demixing. This phenomenon is due, in our case, to the difference between the ionization potential of fluorine and that of sulfur. In an arc discharge the neutral atoms diffuse in from the edges to the axis whereas the ions and the electrons diffuse out from the axis to the edges (ambipolar diffusion). In our experimental conditions the positive ions are predominantly S^+ ions which tend to diffuse to the edges. So, near the axis of the plasma, the relative concentration of sulfur (atoms + ions) is lower than the equilibrium concentration. The main effects of this demixing phenomenon on the axis of a SF_6 arc are the following:

the ratio (called the demixing factor) between the fluorine concentration (atoms + ions) and the sulfur concentration is greater than 6 (6 being the stoichiometric value);

as the electrons are mainly created by sulfur ionization, the real value of the electron number density is lower than the equilibrium value deduced from the electron temperature;

when the discharge current increases, the temperature gradients and thus the demixing effects increase, as long as fluorine is not strongly ionized. For example, with an axial temperature T_0 of 10 000 K we experimentally determined a demixing factor of 7; this value is 8.4 when $T_0 = 11\,000$ K.

Although, at the working temperatures chosen ($T < 11\,000$ K), departures from LTE are slight, they are still taken into consideration during the present study, especially for the determination of the density of F^-. So, knowing N_{S^+}, N_e, and ξ, it is possible to calculate ε_{rec} between 200 and 500 nm.

VI. BREMSSTRAHLUNG

Over the wavelength range which we have studied, the electron-ion bremsstrahlung has an intensity comparable to that of the electron-atom bremsstrahlung:

$$\varepsilon_{brems} = \varepsilon_{e\text{-}i}(\lambda, T) + \varepsilon_{e\text{-}a}(\lambda, T) \ .$$

A. Electron-ion bremsstrahlung

The emission coefficient $\varepsilon_{e\text{-}i}(\lambda, T)$ due to the bremsstrahlung of an ion of charge z is given by Cabannes and Chapelle[18] and is expressed as

$$\varepsilon_{e\text{-}i}(\lambda, T) = C_2 z^2 \frac{N_e N_i}{\lambda^2 T^{1/2}} \exp\left[\frac{-hc}{\lambda k T}\right] G_z(\lambda, T) \ . \quad (3)$$

The factor $G_z(\lambda, T)$ takes into account the averaging over a Maxwellian electron distribution

$$G_z(\lambda, T) = \int_0^\infty g_{e\text{-}i}(y, \lambda, T) e^{-y} dy \ , \quad (4)$$

where

$$y = \frac{1}{kT}\left[\frac{m v_i^2}{2} - h\nu\right] = \frac{m v_f^2}{2kT} \ ,$$

$$C_2 = C_1 \ .$$

v_i and v_f are the initial and final electron velocities. The free-free Gaunt factor $g_{e\text{-}i}(y, \lambda, T)$ is defined by the quantum corrections to the semiclassical calculation.[19]

B. Electron-atom bremsstrahlung

An estimation of the spectral emission coefficient $\varepsilon_{e\text{-}a}(\lambda, T)$ of the electron-neutral free-free radiation is obtained by setting the hypothesis that collisions of electrons with neutral atoms are of the rigid-sphere type. We have[18]

$$\varepsilon_{e\text{-}a}(\lambda, T) = C_3 \frac{N_e N_a}{\lambda^2} T^{3/2} \exp\left[\frac{-hc}{\lambda k T}\right] g(\lambda, T) \ , \quad (5)$$

$$g(\lambda, T) = \int_0^\infty s(y + x_0)(y + x_0)^2 e^{-y} dy \ , \quad (6)$$

with

$$x_0 = \frac{hc}{\lambda k T} \ ,$$

$$y = \tfrac{1}{2} m v_i^2 - x_0 \ ,$$

$$C_3 = \frac{32}{3c^2}\left[\frac{e^2}{4\pi\varepsilon_0}\right]\left[\frac{k}{2\pi m}\right]^{3/2}$$

$$= 1.026 \times 10^{-34} \ \text{(in SI units)} \ .$$

The function $s(y + x_0) = s(E_e)$ is the cross section of electron—neutral-atom collisions. These cross sections are given for F and S by Robinson and Geltman.[5]

The contribution of the total signal arising from the bremsstrahlung is very low (Fig. 5) especially in the high-energy part of the studied spectrum. It was, however, taken into consideration in the evaluation of the total continuum.

VII. S^- ION AFFINITY CONTINUUM

The ground state of the S^- ion is a doublet ($^2P_{3/2,1/2}$). The ground state of the S atom is a triplet ($^3P_{2,1,0}$). The six photodetachment transitions are those which link the two states of the S^- doublet with the three states of the S

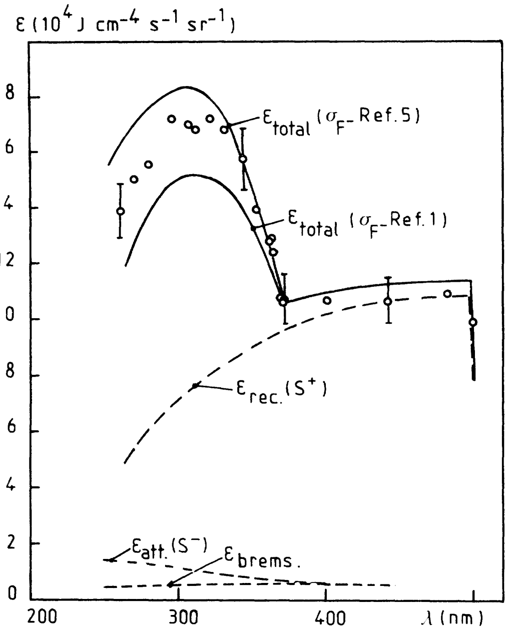

FIG. 5. Reconstitution of the total continuum emitted by a SF_6 plasma. ($T=10\,000$ K; $N_e=2.8\times10^{16}$ cm^{-3}). Comparison with experimental results.

triplet. For the S attachment continuum, Lineberger and Woodward[20] demonstrated the existence of four thresholds, the first being situated at 614.4 nm. This continuum is therefore within the spectral range studied. In the experimental spectrum we were unable to show the presence of the thresholds reported in Ref. 20 as the intensity of ε_{S^-} is weak compared to that of the total continuum. We calculated ε_{S^-} using the photodetachment cross sections proposed by Conneely *et al.*[21] and also those of Robinson and Geltman.[5] It will be seen that the choice of these cross sections does not introduce any notable difference in the final result.

In order to be able to calculate ε_{S^-}, the density of the S^- ions must also be known. From experimental data we therefore calculated the composition of the plasma which allowed us to find N_{S^-} and also N_{F^-}. Note that the inaccuracies of the cross sections σ_{S^-} will only give rise to a very small error in the final result since the negative-ion affinity continuum ε_{S^-} only acts as a correcting term.

VIII. COMPOSITION OF THE PLASMA AND RECONSTITUTION OF THE TOTAL CONTINUUM

It has been seen that the electron density is obtained independently of any hypothesis concerning local thermodynamic equilibrium. Similarly, the temperature T was obtained with Boltzmann's and Saha's laws which only require statistical equilibrium of the species S and S^+. It was also shown that this was reached in our experimental conditions.[15] Furthermore, demixing was taken into consideration. The underpopulation of electrons along the discharge axis, with respect to the equilibrium electron density, is clearly visible in Fig. 4. It should be remembered that the method used for the determination of T allows calculation of the demixing factor, defined in Sec. V. Two other remarks should be made. The demixing phenomenon especially influences the density of sulfur. Considering the stoichiometric ratio between F and S (equal to 6), its influence on n_F is not very important. This is favorable to the accuracy of the measurements. The second remark concerns the change of N_{S^-} and N_{F^-} with temperature. Over the temperature range studied, the densities of the S^- and F^- ions are not very sensitive to changes in T. This is also a favorable element.

A theoretical reconstitution of the total continuum is presented in Fig. 5, where are shown the changes of the various elements of the continuum with λ for a value of T of 10 000 K. For this reconstitution the value of the electron number density ($n_e=2.8\times10^{16}$ cm^{-3}) corresponds to measurements with an arc current of 30 A. The values of the photodetachment cross section of the negative ion S^- are those of Conneely *et al.*[21] Concerning the emissivity ε_{att} due to F attachment, in Fig. 5 we present the two curves deduced from the values of σ_{F^-} given by Mandl[1] and Robinson and Geltman,[5] respectively. The points are the experimental values of ε_{tot} measured along the axis of a 30-A discharge, with corresponding error bars.

IX. CROSS SECTION OF F^- PHOTODETACHMENT

Figure 6 gives the variation of σ_{F^-} versus the wavelength λ. The same figure gives the experimental values of Mandl[1] and the theoretical values of Robinson and Geltman,[5] Clodius *et al.*,[8] Ishihara and Foster,[6] and Rescigno *et al.*[7] The cross section σ_{F^-} is obtained from formula (1) which, using Saha's law, can be written

$$\sigma_{F^-}=C\frac{\varepsilon_{att}(\lambda,T)}{N_eN_FT^{3/2}\exp\left[\dfrac{-hc}{\lambda kT}+\dfrac{E_I}{kT}\right]}, \tag{7}$$

where E_I is the electron affinity.

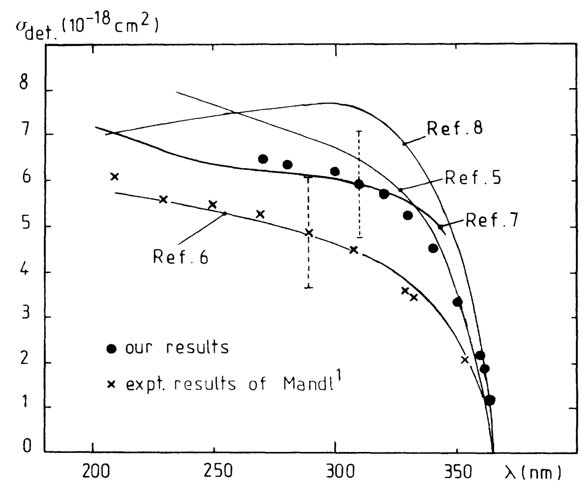

FIG. 6. Fluorine photodetachment cross sections. Comparison of different theories and experiments.

The error introduced to the total continuum ε is evaluated at 7% and the error of ε_{att} is lower than 10%. The uncertainty of N_e is of the order of 8%. We checked that the value of N_e measured from the recombination continuum was the same as that measured by interferometry. To minimize the error of ε_{att} near the threshold, the value of N_e selected (within the error range) was therefore one which gives a recombination continuum (for $\lambda \simeq 370$ nm) exactly equal to that determined experimentally. For this we took into account the continuum arising from bremsstrahlung and from the S^- ions. An iterative method was used making the assumption firstly, that the total continuum for $\lambda = 370$ nm was entirely due to the recombination of the S^+ ions ($N_e = N_{S^+}$). Then, for the value of T corresponding to the experimental conditions, we calculated ε_{brems} and ε_{S^-} which we subtracted from ε_{total}. From the recombination continuum thus deduced by means of formula (2), a new value can be obtained for N_e which is substituted in the calculations until the system converges towards a final value of N_e (two iterations are sufficient). Note that with the cross sections of photodetachment of S^- proposed by Conneely *et al.*[21] the initial density N_e was 2.99×10^{16} cm^{-3} giving a convergence value of 2.81×10^{16} cm^{-3}. If in the calculation of σ_{S^-} the values of Robinson and Geltman[5] are used for the same initial density, the convergence value is 2.72×10^{16} cm^{-3}. For the final result of σ_{F^-}, the difference obtained in the rise of one or other of the cross sections for S^- is lower than 3%.

The choice of the value of N_e means that the error in ε_{att} near the threshold is lower than 5%. Likewise, the error in the determination of N_F is low because N_F varies slowly with T. Moreover we mentioned that demixing effect only plays a minor part in the density of neutral F. A further cause of error could be a difference between the electron temperature T_e and the temperature of the heavy particles T_g. We have shown elsewhere[15] that the difference $T_e - T_g$ is of the order of 300 K in our experimental conditions. As Dalton's law is used to calculate the composition, this difference gives rise to an underestimation of N_F of about 3%. So we increased the values obtained for N_F, assuming LTE, by 3%. In these conditions the N_F values have an error of less than 4%.

The error of the last term, which contains the temperature, varies with the wavelength. It is highest around the threshold where it is estimated to be at most 4.5%.

The total maximum error possible is therefore estimated to be 24%.

Our values are in good agreement with the theoretical values of Robinson and Geltman[5] and with those of Rescigno *et al.*[7] They are intermediate between the values of Clodius *et al.*[8] and those of Ishihara and Foster.[6] Considering the error margins, they are not in disagreement with Mandl's experimental values[1] but are about 25% higher in all cases.

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