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Electron detachment and excitation processes in F^- -inert-gas collisions

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Abstract. The results of a study of electron detachment processes occurring in F^- -inert-gas collisions in the 10 eV-4 keV laboratory collision energy range are reported. A differential time-of-flight energy loss study of scattered F atoms and an electron spectroscopy study were performed. The differential cross sections for the various processes observed are reported. As in other halogen negative-ion collisions, 'dynamic' effects in the direct detachment process were observed. The relative importance of excitation processes was determined. The production of a series of autodetaching and autoionising states is reported.

1. Introduction

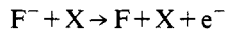
In this work we present the results of an experimental study of collisions between F^- and several inert-gas (IG) targets in the low to medium energy range, extending from tens of eV to 4 keV. Our work extends previous studies of detachment in other halogen negative-ion-inert-gas collisions (Cl^- , Fayeton *et al* 1978, Br^- , I^- , Esaulov 1981 and references therein). This work was motivated by an absence of detailed theoretical studies of detachment for the case of negative ions with $l \neq 0$ outer electrons. For these cases, at low energies where a molecular description should be valid, models such as Demkov's (1964) zero-range potential model may not be applicable *a priori*. Other models can be envisaged, such as a short-range potential model (see, for example, Drukarev 1981, Gauyacq 1983) which circumvents the inadequacies of the zero-range description, or the rotational coupling model suggested by De Vreugd *et al* (1982). None of these have yet been applied. It was thus thought that thorough investigation of collisions involving the simplest halogen anion, F^- , would be useful, since they could provide a 'simple' object for a theoretical investigation.

The present study complements some recent total-detachment measurements (Huq *et al* 1982) for collisions between F^- and either He, Ne or Ar for energies below 300 eV, and some summed neutral and summed ionic differential cross section measurements of De Vreugd *et al* (1982). In order to obtain a comprehensive overview of detachment two complementary techniques were employed. A differential time-of-flight (TOF)

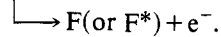
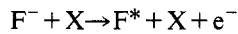
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energy loss study was performed in order to study separately the various detachment channels, for example:

direct detachment (DD)



detachment with excitation (DE)

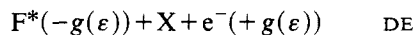
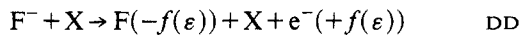


This study was complemented by measurements of the detached-electron energy spectra (DEES), which give information about both direct detachment and the production of autoionising and autodetaching states and their decay channels.

2. Experiment

The experimental set-ups and methods used have been previously described (Esaulov *et al* 1978, time-of-flight; Montmagnon *et al* 1983, electron spectroscopy) and we shall only mention the principal characteristics of the measurements.

Time-of-flight measurements were performed in the 300 eV–2 keV energy range. The energy resolution in the incident ion beam depends on the collision energy and is typically 0.8 eV at 1 keV. The actual resolving power in so far as the identification of excitation processes is concerned is, however, determined by electron ejection. Indeed, by energy conservation the energy loss spectrum of the F atom reproduces that of the detached electron:



where $f(\varepsilon)$ and $g(\varepsilon)$ are the energy (ε) distribution of the detached electron.

This provides a very convenient method of studying detached-electron energy spectra in the DD channel as a function of impact parameter (since a study, differential in angle, is performed), but in the case of excitation processes makes an unambiguous identification of excitation processes difficult. Electron ejection kinematics also affects the position of energy loss peaks (see Esaulov *et al* 1978). The angular resolution was typically 0.12° . Differential cross sections (DCS) were obtained by integrating the characteristic peaks in the neutral energy loss spectra: the results are reported as plots of the reduced DCS: $\rho(\tau) = \sigma(\theta)\theta \sin \theta$ against $\tau = E\theta$.

Detached-electron energy spectra (DEES) were measured in the energy range from 10 eV to 4 keV for a 90° observation angle. In some cases, when identification of autodetaching or autoionising states had to be made, angular measurements were performed in order to make use of ejection kinematics. The energy resolution in these measurements was typically 50 meV. The calibration of the electron energy was made against the resonance structure in O_2 (Esaulov *et al* 1984).

3. Results and discussion

3.1. Energy loss spectra

Figure 1 shows some typical neutral energy loss spectra. Note that the zero of the energy loss scale was taken so that the elastic scattering F⁻ peak appeared at -3.4 eV, the F⁻ binding energy. For all the targets investigated (He, Ne and Ar) the spectra consisted of two peaks. The full spectrum with the two peaks is shown only for one scattering angle for simplicity.

The first peak (DD) corresponds to the direct-detachment process. This peak is fairly large and broadens as the scattering angle increases (see figures 1(a), (b) and (d)). As mentioned above this peak reproduces the detached-electron energy spectrum for a given impact parameter. One can thus conclude that in all the studied cases the DEES broaden with decreasing impact parameter. This dependence has also been encountered in H⁻-IG collisions (Vu Ngoc Tuan *et al* 1983) and is also known in ionisation collisions (for example, in He⁺+He (Barat *et al* 1972, Sidis 1973)) and was well accounted for by theory. It is interesting to note, however, that such a dependence was *not* observed for other halogen negative ions (Esaulov 1981) where the DEES has been found to become *narrower* as the impact parameter decreases. The difference between the dependences observed for F⁻ and other halogen anions is not clear. The narrowing of the spectra as the impact parameter decreases seems difficult to account for. Such a dependence is predicted in the sudden approximation ZRP model of Bronfin and Ermolaev (1971) but it has not yet been shown that the premises of this model correspond to a realistic description of the collision.

The second peak DE corresponds to detachment accompanied by excitation processes involving either F (or F⁻) excitation or target excitation.

3.1.1. F⁻-He. As can be seen in figure 1(a), in F⁻-He collisions only F excitation is observed. No clear indication of He excitation is present. This spectrum shows that the low-lying members of the F* (2p⁴(³P)nl) or F^{-*} (2p⁴(³P)nlnl') series are not excited. This is not surprising since, in the absence of target excitation, this would involve the breakdown of the spin conservation rule. Excitation processes thus involve mainly F* (2p⁴(¹D)nl) or F^{-*} (2p⁴(¹D)nlnl') excited states. This conclusion is in agreement with earlier electron spectroscopy measurements of Edwards and Cunningham (1974) where only the F^{-*} (2p⁴(¹D)3s²)¹D state lying at 14.85 eV was observed. In the TOF spectrum the energy loss corresponding to DE is greater than 15 eV; thus the excitation of the F⁻ state is relatively small and it is the F* (2p⁴(¹D)3s) state that dominates.

3.1.2. F⁻-Ne. In the case of the quasisymmetric F⁻-Ne the assignment of excitation processes is difficult (figure 1(b)). As for F⁻-He collisions excitation of F* (2p⁴(¹D)nl) or F^{-*} (2p⁴(¹D)nlnl') states appears to be dominant. However, contrary to the F⁻-He case, excitation of the 2p⁴(¹D)3s state seems less important. Ne excitation could also be present. As is mentioned below, electron spectroscopy measurements did not reveal any excitation of Ne^{-*} autodetaching states. Only F⁻ states were observed.

A significant cross section for F⁺ production was observed (see below).

3.1.3. F⁻-Ar. In the case of the Ar target, figure 1(c) shows that mainly Ar or Ar⁻ is excited.

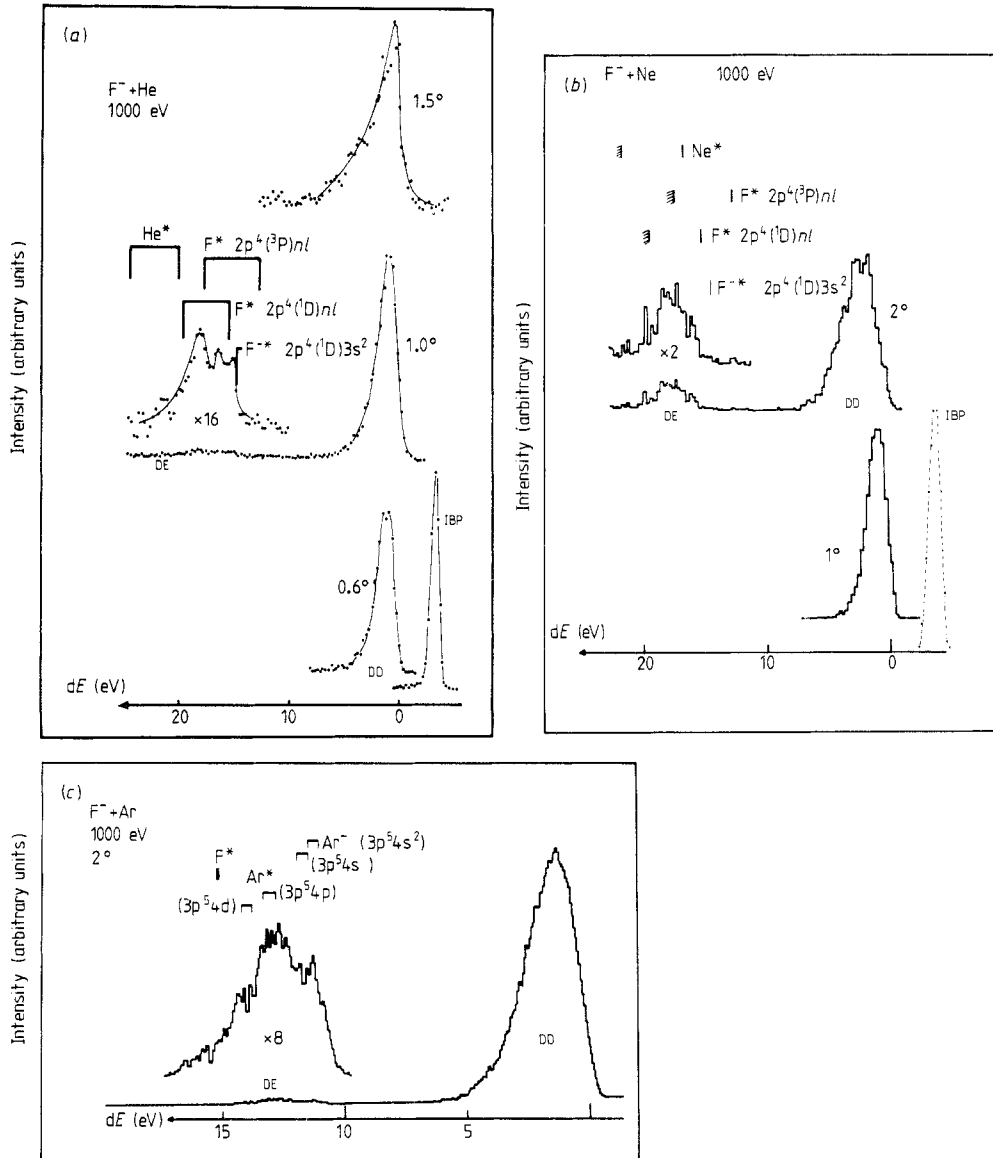


Figure 1.

3.2. Differential cross sections (DCS)

The measured DCS are shown in figure 2. The general form of all the neutral DCS remains unchanged in the studied energy range. The DCS corresponding to direct detachment increase rapidly in the 0.5–1 keV deg angular range, reach a broad maximum and then decrease for larger τ where the DCS for the excitation processes becomes important. This behaviour is quite common in negative-ion-atom collisions and has been observed in our previous studies (H^- -IG collisions (Esaulov *et al* 1978) and halogen negative-ion collisions (Esaulov 1981)). It may be tempting to ascribe the peaked shape in the DD DCS to a localised direct detachment process. However,

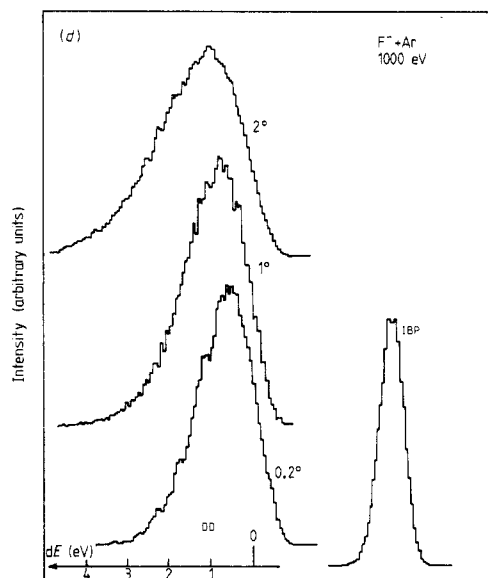


Figure 1. TOF neutral energy loss spectra for several scattering angles. Laboratory energy = 1000 eV. The ion beam profile (IBP) is also shown. The line which is drawn through the experimental points in figure 1(a) is a guide for the eyes. (a) F⁻ + He (0.6°, 1.0°, 1.5°); (b) F⁻ + Ne (1.0°, 2.0°); (c) F⁻ + Ar (2.0°); (d) F⁻ + Ar (0.2°, 1.0°, 2.0°).

such a conclusion made on the face of the DCS would not in general be true. Indeed, in the case of H⁻-He collisions the fall off of the DD DCS for large τ was related to the characteristics of elastic and inelastic scattering in the parent HHe system (see Esaulov *et al* 1978), whereas the detachment probability remained essentially constant for large values of τ .

Our results are in good agreement with the summed neutral DCS measured by De Vreugd *et al* (1982). It should be noted that in the case of F⁻-Ne collisions the summed ionic cross section measured by De Vreugd *et al* (1982) represents the sum of the F⁻ and F⁺ cross sections. In F⁻-Ne collisions, F⁺ production is important, contrary to the cases of other targets, where we did not observe any F⁺. This is also the case in Cl⁻-IG collisions (see Fayteon *et al* 1978) where Cl⁺ was only observed in Cl⁻-Ar collisions.

In order to estimate the importance of the different detachment channels the DCS were integrated over scattering angles for a 2 keV collision energy. It was thus found that detachment with excitation represents $(16 \pm 1)\%$ of the total detachment cross section σ_{-10} in F⁻-He collisions and $(17 \pm 1)\%$ and $(14 \pm 1)\%$ in F⁻-Ne and F⁻-Ar collisions respectively. Assuming *equal* detector efficiencies for F⁺ ions and F atoms at this energy we find that, in the angular range of the present measurements, F⁺ production represents $(23 \pm 1)\%$ of the total electron production cross section $(\sigma_{-10} + \sigma_{-11})$ in F⁻-Ne collisions.

The neutral DCS considered do not give any direct indication of the dynamics of the detachment process. A study of the energy dependence of the elastic DCS is useful in this case. Indeed it may be shown (see Lehman and Liebfried 1962) that, for relatively high energies and small scattering angles, the elastic DCS scales for different collision energies when plotted in a $\rho(\tau)$ plot. This is true in the absence of inelastic processes. This scaling does not persist beyond τ values corresponding to the onset of some inelastic channel, which will manifest itself as an absorption of the reduced elastic DCS. The study of this absorption as a function of τ for different collision energies can give some insight into the dynamics of the process. Figure 3 shows this

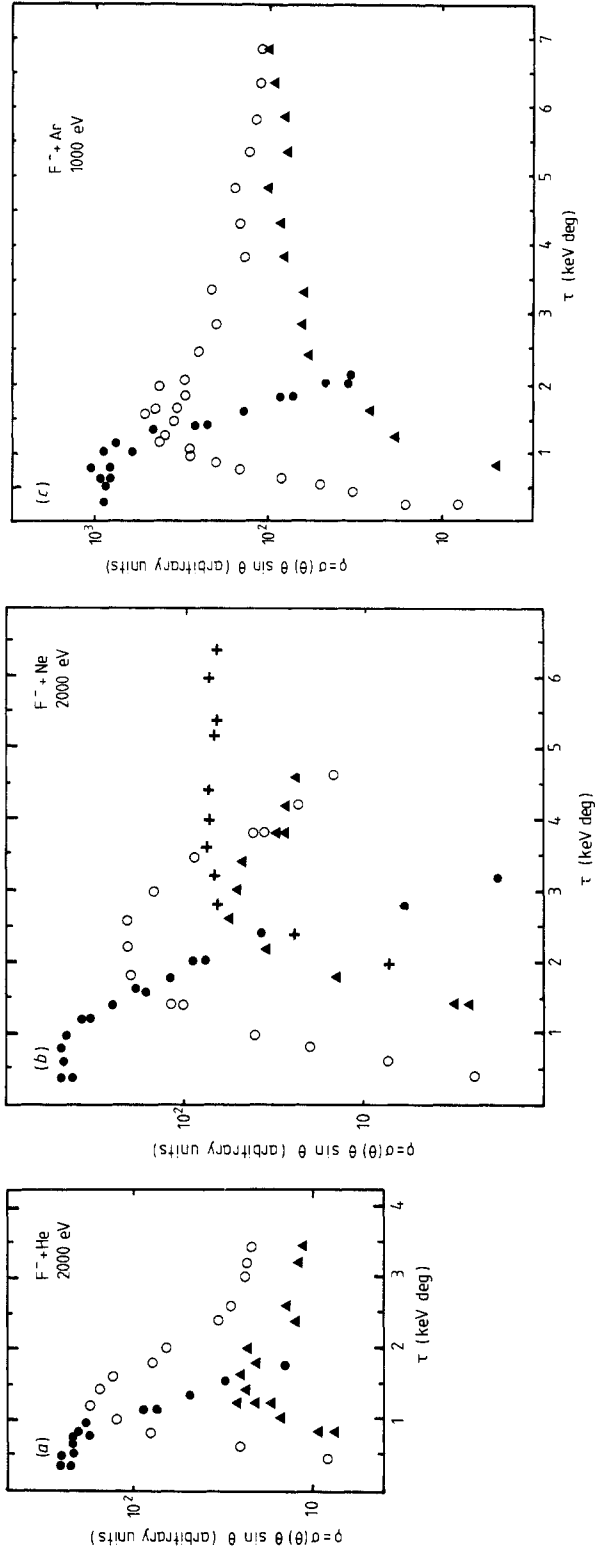


Figure 2. Reduced differential cross sections: ●, elastic; ○, DD, direct detachment; ▲, DE, detachment with excitation; ⊕, production of F⁺. (a) F⁻ + He (2000 eV); (b) F⁻ + Ne (2000 eV); (c) F⁻ + Ar (1000 eV).

dependence in the case of F⁻-He collisions at 300 and 750 eV. As can be seen, the absorption of the elastic DCS increases with increasing collision energy. This behaviour is contrary to that expected in the local complex-potential model of detachment, where it is the time spent in the continuum by the system which plays an important role and the detachment probability is larger for smaller velocities for a given impact parameter or τ . The increase of absorption in the elastic DCS that we observe is related to the increase of DD with increasing collision velocity and also to the increase in the importance of DE. Figure 2(a) shows that, for example, at 1 keV deg the DCS for the DE process represents about 0.1 of the DD DCS for a 2 keV collision energy. On the other hand, at 1 keV (see figure 1(a)) it may be seen that this ratio falls to about 0.04. Thus, for energies below 1 keV, excitation processes are not significant. This result is consistent with electron spectroscopy measurements (see below) which show that excitation processes are first clearly visible at a 1 keV collision energy. It thus appears reasonable to assert that the rather large increase in the absorption of the elastic cross section as the energy increases from 300 to 750 eV is in fact due to direct detachment. Hence in F⁻-He collisions the direct-detachment mechanism has a 'dynamic' character, i.e. is affected by the increase of internuclear *velocity* and is not 'static' as in the local complex-potential model, where the imaginary part of the potential is obtained without any regard to nuclear motion.

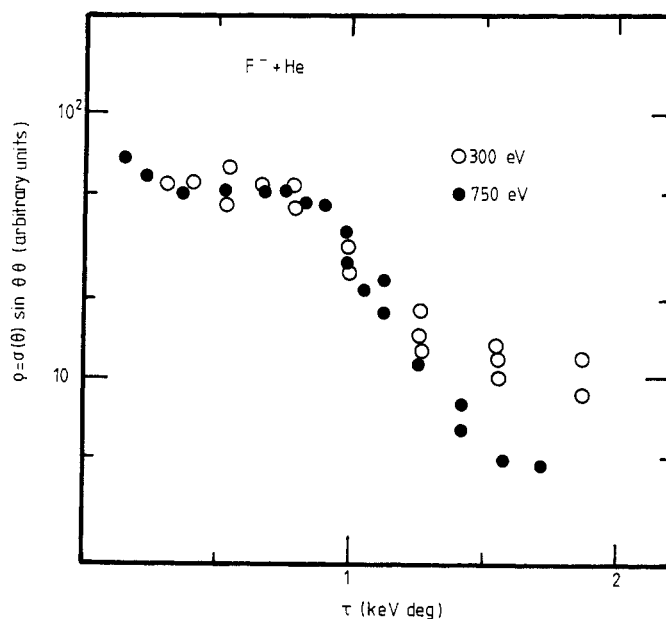


Figure 3. Reduced elastic differential cross sections for F⁻+He at different laboratory energies: ○, 300 eV; ●, 750 eV.

3.3. Electron spectroscopy

Figures 4-7 show some results of the direct measurement of DEES which complement the TOF study. As noted previously, these measurements correspond to a 90° observation angle with respect to the incident beam direction. It should also be borne in mind that these DEES represent sums over all impact parameters and all detachment channels.

3.3.1. F^- -He and F^- -Ne. Figure 4(a) shows the DEES obtained in F^- -He collisions. As the collision energy increases the spectra are found to broaden out. For energies above 500 eV the DEES display well defined structures. The position of these was found to change as a function of the observation angle and the shift in energy was consistent with what is expected on the basis of electron ejection kinematics if F^- is the emitter. It was thus concluded that these structures are due to the decay of autodetaching F^- states or autoionising states of F. Measurements of DEES for higher electron energies did not reveal any structure other than the peak due to the decay of the $2p^4(^1D)3s^2 F^-$ resonance previously reported by Edwards and Cunningham (1974).

Similar observations were made in the case of Ne (figure 4(b)). The DEES at low energies are structureless and are found to broaden out as the collision energy increases. For energies above about 100 eV most structures observed in F^- -He collisions appear in the spectra. The high-energy DEES did not reveal any structure other than the one corresponding to the $2p^4(^1D)3s^2$ state of F^- . A careful search for any Ne^- autodetaching

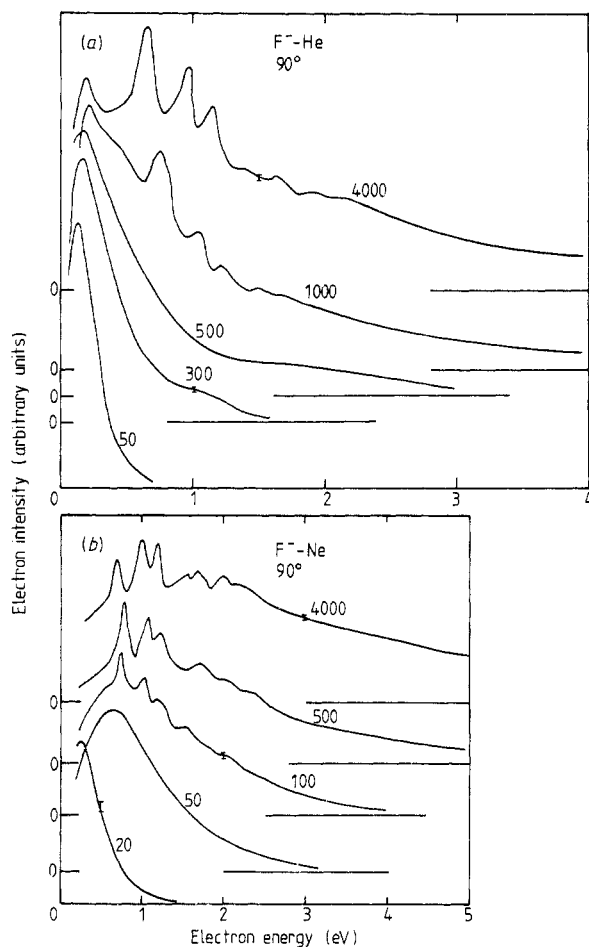


Figure 4. Detached-electron energy spectra corrected for analyser transmission at different laboratory energies. (a) $F^- + He$ (50, 200, 500, 1000, 4000 eV); (b) $F^- + Ne$ (20, 50, 100, 500, 4000 eV).

states was also made but none were observed. All autodetaching states excited thus correspond to F⁻.

No sign of autodetaching states corresponding to the 2p⁴(³P)3s² configuration, which should decay into the ground state, was observed for either the He or the Ne target, in agreement with what is expected from the spin conservation rule when the target is not excited.

3.3.2. F⁻-Ar, Kr and Xe collisions. The measured DEES are shown in figure 5. In this case no low-lying structures were observed. The spectra are smooth and broaden out as the collision energy increases.

The study of DEES for electron energies above 5 eV (figure 6) reveals a fairly rich structure. For all these systems no F⁻ excitation is observed and all the peaks in the electron spectra correspond to either autodetaching states of the inert-gas anion or to autoionising states of the inert gas.

In all cases the main excitation process corresponds to the production of the np⁵(n+1)s² (inert-gas) resonance, well known from electron spectroscopy studies (see Schulz 1973). A statistical ratio for the population of the Ar⁻ ²P_{3/2} and ²P_{1/2} states is reached at the 4 keV collision energy. At lower energies this ratio is quite different, as is demonstrated by the inset in figure 6(a). At low collision energies this change and the modification of the form of the peaks may be related to molecular effects, i.e. decay during the collision. Since no information about the lifetime of the quasimolecular complex exists, no definite statement can be made at present. Note that some modification (broadening) of the peaks will also occur related to kinematic effects due to the elastic energy gain of Ar during the collision. In the case of Kr and Xe the population of the ²P_{1/2} states remained quite small.

The electron spectra also display other structures lying at lower and higher energies. These may correspond to both autodetaching states or autoionising states of the inert gas. A comparison of these spectra with the ones observed in He-IG collisions by Gerber *et al* (1972), where mainly doubly excited autoionising states were identified, shows a strong similarity. However, here only a few lines in the case of the Kr and Xe targets are well resolved. Tentative attributions are given in figure 6.

3.3.3. Fine structures in F⁻-He and F⁻-Ne DEES. Figure 7 shows a high-resolution study of the low-energy DEES for a 4 keV collision energy and a 30° observation angle for F⁻-He and F⁻-Ne collisions. The spectra display a rich structure. Most peaks are observed for both the He and Ne targets. In both F⁻-He and F⁻-Ne collisions the peaks (E, F), (H, I) and (J, K) appeared at the lowest collision energies. The peak (B, C), not seen in the 4 keV F⁻-Ne spectrum, appears as a small structure at lower (500 eV) energies. A general feature of most of these peaks is the more or less well defined splitting into two components with a separation of roughly 40 meV. The energies of these lines in the emitter frame of reference are tabulated in table 1.

The observed lines may be due to the decay of autodetaching states of F⁻ or autoionising states of F. Edwards and Cunningham (1974) have reported the observation of autoionising states of F in their study of 4 keV F-He collisions. The lines observed were assigned to excited states of F of the 2p⁴(¹D)nl type lying above the F⁺ (³P_{2,1,0}) continuum, which are hence unstable. Some of these states were first identified in a photoabsorption experiment by Huffman *et al* (see Edwards and Cunningham 1974). Edwards and Cunningham also report peaks from the decay of states of the 2p⁴(¹S)nl configurations into the F⁺ (¹D) continuum. Decay into the F⁺ (³P_{2,1,0}) continuum will generate triple peaks with a 0.0423 eV (³P₂-³P₁) and 0.185 eV (³P₁-³P₀) splitting.

The results of Huffman *et al* and Edwards and Cunningham (1974) as well as the suggested assignments are listed in table 1 along with our data. Most of the lines are seen to originate from autoionising states of F, although some ambiguities concerning peak locations do exist. These are partly due to uncertainties in both the present data and those of Edwards and Cunningham (1974) (± 30 meV and ± 50 meV respectively). The doublet splitting of about 40 meV in figure 7 (see brackets in table 1) is due to

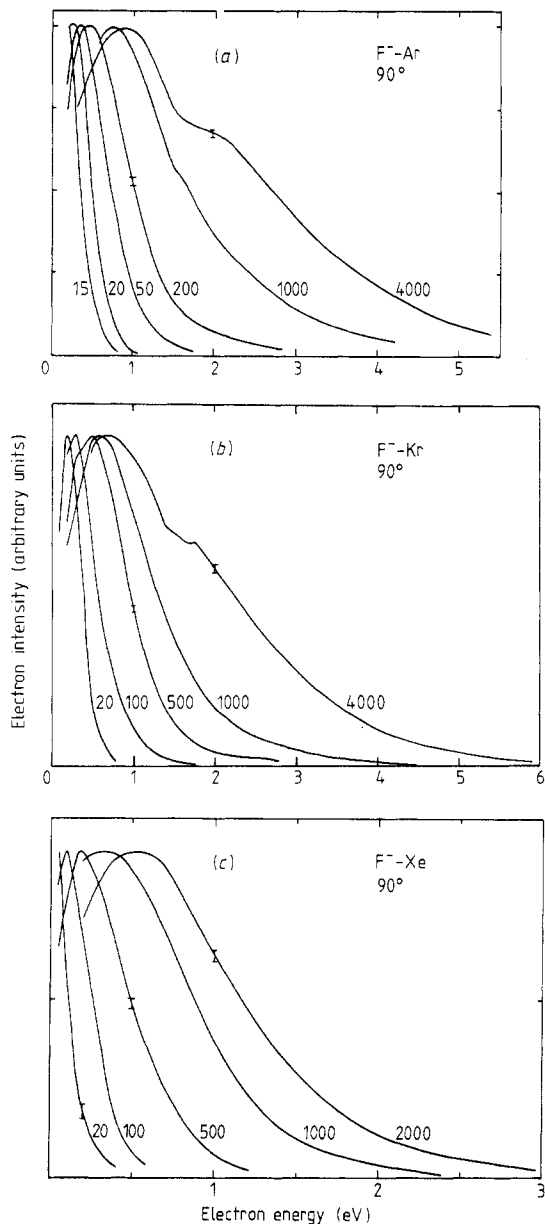


Figure 5. Detached-electron energy spectra corrected for analyser transmission at different laboratory energies. (a) $F^- + Ar$ (15, 20, 50, 200, 1000, 4000 eV); (b) $F^- + Kr$ (20, 100, 500, 1000, 4000 eV); (c) $F^- + Xe$ (20, 100, 500, 1000, 2000 eV).

Table 1. Energy of peaks due to the decay of autoionising states of F 2p⁴(¹D)nl' and 2p⁴(¹S)nl'. Some of the peaks were first identified by Huffman *et al* and Edwards and Cunningham (1974) (EC).

Peak	Energy (eV)			
	Present work	(EC)	Huffman	F*
A	0.093	0.10		3p''
B	0.224			
C	0.256			
D	0.49			
E	0.711			} 4s'
F	0.745	0.76	0.764	
G	0.868	0.87		4s''
H	1.00			
I	1.030	1.06	1.046	} 3d'
			1.069	
J	1.179			} 4p'
K	1.210			
L	1.24			
M	1.38			
N	1.44			} 3d''
		1.49		
O	1.559			} 5s'
P	1.606	1.60	1.615	
		1.66		4p''
Q	1.69			} 4d'
R	1.725		1.725	
			1.735	
S	1.810			5p'
T	1.98	2.00	1.984	6s'
U	2.03			5d'
V	2.07	2.08		6p'
W	2.19			} 6d', 5p''
X	2.30			

decay into the ³P₂ and ³P₁ states of F⁺. The ratio of the peak heights is found to be approximately equal to the statistical ratio of 1.66 (³P₂/³P₁). Our experimental resolution would not have been sufficient to resolve peaks due to decay into the ³P₁ and ³P₀ states. Note also that the statistical weight for decay into the ³P₀ state is small, being a third of that for decay into the ³P₁ state.

Some of the peaks, like (B, C), cannot be identified in this scheme. These may result from the decay of a F^{-*} state to an excited F state. A possible candidate for the (B, C) peaks would be a transition between the F⁻ (¹D)3s3p ³P and F (¹D)3p states. This proposal is based on an estimation of the F⁻-state energies using the modified Rydberg formula of Read (1977). Other transitions of this type may also contribute to our observed spectra.

Alternatively the A, B, C peaks could be due to the decay of F^{-*} (¹D)3s² states, lying at 14.85 eV, into *excited* states such as the F* 2p⁴(³P)3p ²P (peak A) or F* 2p⁴(³P)3p ²D (peaks B and C). The latter explanation, suggested by Clark *et al* (1985), could also explain the smallness of these peaks in F⁻-Ne collisions because of the quasisymmetry consideration discussed above.

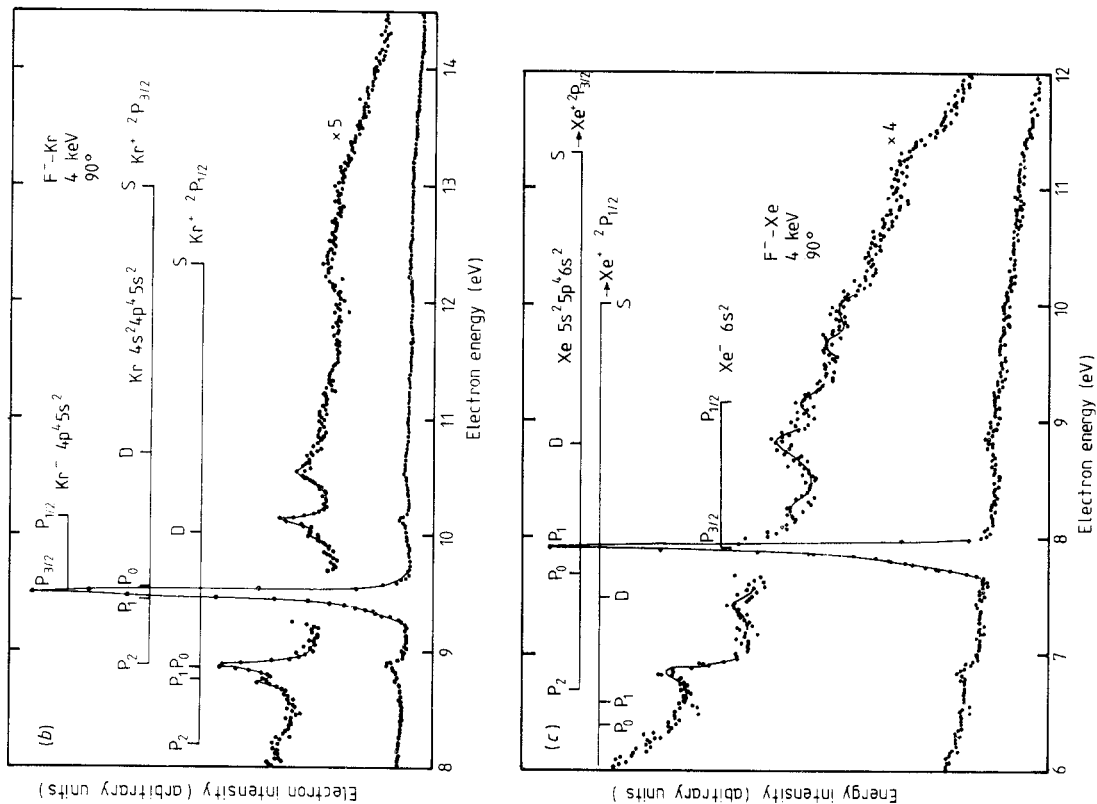


Figure 6. Detached-electron energy spectra in the 10-20 eV range at 4 keV. (a) $F^- + Ar$; (b) $F^- + Kr$; (c) $F^- + Xe$.

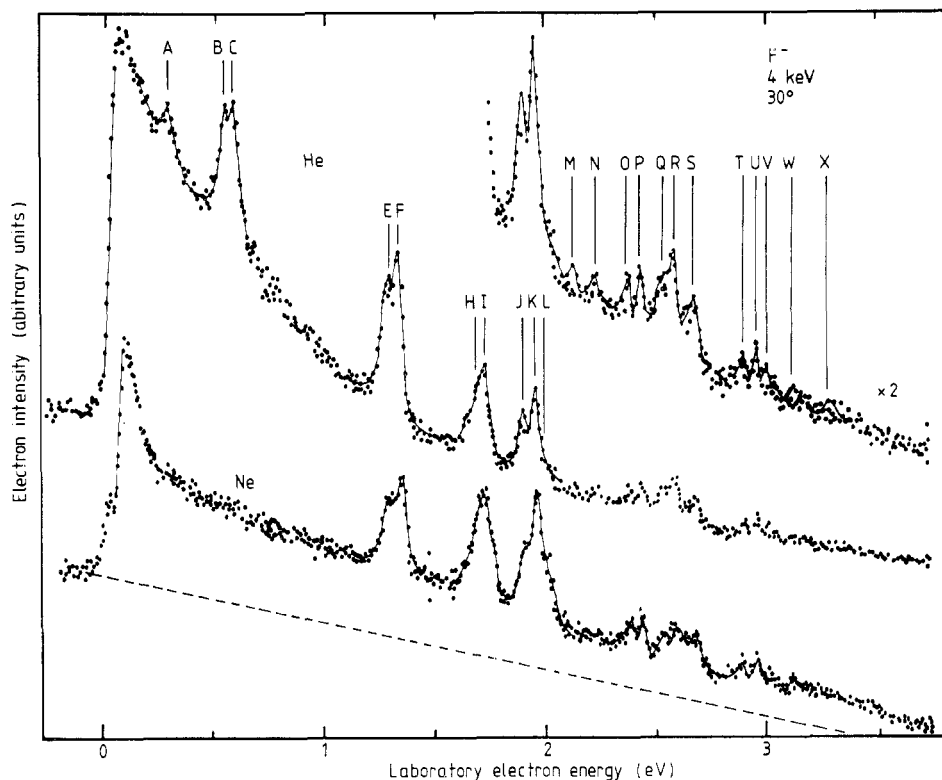


Figure 7. High-resolution detached-electron energy spectra of F⁻ + He and F⁻ + Ne at 4 keV.

4. Concluding remarks

Our paper presents some detailed studies of direct electron detachment and detachment accompanied by excitation processes in F⁻-inert-gas collisions.

Our results show that, as in other negative-ion systems (H⁻, and some other halogen-anion-inert-gas collisions) electron detachment has a 'dynamic' character. Thus in F⁻-He collisions the detachment probability is found to increase with collision velocity contrary to what may be expected in a local complex-potential model. The F⁻-He system is the simplest halogen-anion-inert-gas system and it is hoped that the present results and previous total cross section measurements of Huq *et al* (1982) will stimulate a detailed *ab initio* investigation of detachment in this system.

No attempt was made in this paper to discuss at length, at least in qualitative terms, the observed excitation processes. In a previous paper on Cl⁻-IG collisions, Fayeton *et al* (1978) have discussed excitation processes in terms of the MO model (see, for example, Barat and Lichten 1972). The present results for F⁻ are basically in agreement with such a description. Thus, as expected, excitation processes are found to involve F (or F⁻) in F⁻-He and F⁻-Ne collisions and the heavier inert-gas partner is found to be excited in the case of Ar, Kr and Xe. In F⁻-He and F⁻-Ne collisions, excitation of the 2p⁴(¹D)nl states was expected and is observed. By analogy with the isoelectronic Ne-Ne system (see, for example, Gauyacq 1978), quasi-u-g symmetry arguments suggest that the lowest 2p⁴(¹D)3s state should not be significantly excited as is indeed

observed. We have refrained from repeating the discussion of Fayeton *et al* (1978), because it is not clear that an MO description is applicable for a negative-ion collision with a very loosely bound outer electron. Note that a different 'hybrid' model was proposed for H^- by Esaulov *et al* (1978) in which the core ($H-1G$) collision was considered independently of the evolution of the outer electron. Such a description in the case of F^- would lead to much the same results as a direct application of the MO model. If we admit that an MO description is valid in the *early* stages of the collision (while the incoming $X^1\Sigma$ molecular negative state is bound), then one can expect that (see Fayeton *et al* 1978, Esaulov 1980) an outer σ electron will be promoted and detached leading to the population of the $^2\Sigma$ state: $^1\Sigma(\pi^4\sigma^2) \xrightarrow{\sigma} ^2\Sigma(\pi^4\sigma)$. The excitation processes will then be the same as may occur in an F-He collision evolving in only the $^2\Sigma$ state and not as in a 'true' collision in both the $^2\Sigma(\pi^4\sigma)$ and the $^2\Pi(\pi^3\sigma^2)$ state. Our results are also consistent with what may be expected by treating separately the outer electron and the FHe ($^2\Sigma$) core collision, which may be described in a 'standard' MO model. Note that production of autodetaching states in such a model occurs as a result of recapture into an excited state in the outward leg of the collision.

Although our results can be qualitatively understood they do not indicate which model would be appropriate for the excitation process; this should also be the target of detailed theoretical study.

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