Origin of features in the energy spectra of electrons detached from fast H− in collisions with He and Ar atoms

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Abstract. Using Born's approximation we have analysed the structure of the doubly differential cross section for electron loss from fast H− near the forward direction. We show that electrons ejected with velocities slightly smaller than the projectile, originate in collisions in which the 'diffuse' electron of H− scatters as a nearly-free electron and excites (ionises) the target. Electrons ejected with velocities close to that of the projectile come from three sources: (i) double-electron loss in which the final state is strongly influenced by the Coulomb interaction with the resulting fast proton; (ii) single electron loss in which the target is excited by the H core, while the ejected 'diffuse' electron acts as a spectator; and (iii) single detachment with no excitation of the target, which comprises a small fraction of the events for He targets, but becomes important for heavier targets such as Ar.

1. Introduction

The knowledge of electron loss from fast H− in gases has been increased considerably by recent measurements of doubly differential cross sections (DDCS) (Duncan and Menendez 1977, Menendez and Duncan 1979, Macek et al 1984). These results follow closely the expectations of a simple electron scattering model in which the projectile's electron is considered to act as a free electron scattering elastically from the target. In this model, the average velocity of the ejected electron should be equal to that of the projectile, \(v_p\), and the velocity distribution should reflect that of the bound electron state before the collision, modified by the field of the ionised projectile. The main source of discrepancy between experiments and the model is the observation of two peaks in the DDCS: one (\(P_0\)) at \(v_p\) observed only close to the forward direction, and another (\(P_1\)) at a most probable velocity slightly lower than \(v_p\) which has a wider angular spread and conforms better to the distribution expected from the model. The energy of \(P_0\) was found to be approximately \(mv_p^2/2\), independently of the projectile or target. This, together with its shape near zero degrees, resembling that observed for electron capture to the continuum of fast ions, has led to its assignment to double electron loss (DEL) by Duncan and Menendez, with the ejected electrons strongly affected by the Coulomb field of the fast proton. The origin of \(\Delta E\), the difference in electron energy between \(P_0\) and \(P_1\), and the dependence on the target and observation angles, remained unexplained.

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Franz et al (1981) have proposed that both peaks result from single electron loss (SEL) collisions, in which momentum is mainly transferred to the resultant H atom \((P_0)\), or to the ejected electron \((P_1)\). They use a simple description of the electrons of \(H^-\), which for the initial state is represented by a symmetrical product of 1s orbitals. For the final state the ejected electron is approximated by a plane wave \(|k\rangle\), while the resultant H atom is described by an unperturbed 1s orbital; furthermore, this final state is orthogonalised to the proposed initial state.

Day (1982) has studied in detail the effect on the resulting electron distribution of the approximations made on the \(H^-\) wavefunction. He considers a one-electron model in the field of an inert H(1s) core; proposing a 1s orbital for the initial state and a plane wave for the final continuum state without orthogonalisation, he obtains an electron distribution with only one peak. The same happens when the active electron is described in a static potential which is chosen to support one bound state. Finally, considering the two \(H^-\) electrons as active and including radial correlations between them, there results an electron distribution with two peaks symmetrically situated with respect to the projectile velocity \(v_p\).

Maleki and Macek (1982) also consider SEL to describe the electron distribution in H\(^{-}\)-He collisions. They try the simple wavefunctions used by Franz et al but without orthogonalisation, and find a one-peak distribution; the doubly peaked distribution of Franz et al is therefore considered to agree fortuitously with measurements, since the double peak arises only when orthogonalising the initial and final states. However the results obtained by Maleki and Macek are not meaningful, since the projection of the final state on the initial is quite large, resulting in \(\langle k|1s_d\rangle = 7.66\) for \(k = 0\). Finally, they assume a more realistic description for the continuum electron of the final state, using Schwinger’s variational principle for the s partial wave of a continuum orbital in the field of a H(1s) atom; they find its phaseshift to approach the value \(\pi\) at low energy, which is the correct limit for a potential sustaining one bound state. The same formalism is used to obtain an orthogonal initial state. The resulting electron distribution presents two peaks, but its dependence on electron energy and scattering angle is not as good as that of Franz et al when compared with measurements. More recently, Macek et al (1984) have extended these calculations to other targets.

The works of Day and of Maleki and Macek point out the importance of a correct description of the two-electron state of \(H^-\). Nevertheless, the wavefunctions used by Franz et al should not be discarded right away; on the contrary, the bound state is formed by a contracted \(1s_c\) and a diffuse \(1s_d\) orbital of mean radius \(r_c = 0.96\) and \(r_d = 3.57\) au, it presents therefore a strong radial correlation between electrons. The continuum electron of the final state will be poorly described by a plane wave, especially in the neighbourhood of H(1s); nevertheless, in the Born approximation the transition amplitude depends on the overlap between the initial and final states of the active electron, and the region that contributes most to this overlap, where \(r^2 1s_d(r)\) is maximum, is of the order of 7 au. In this region, the plane wave may be adequate to describe continuum electrons, especially those with not too small energies. We will use these states to study SEL and DEL contributions to the ejected-electron distribution for He and Ar targets.

Our aim in this work will be to determine the mechanisms that produce the peaks of the DDCS. We find that peak \(P_1\) is produced by collisions of the weakly bound \(1s_d\) electron of \(H^-\) with the target. As shown by Bell et al (1978), and confirmed by Day and by Maleki and Macek, the detachment collisions of \(H^-\) with He at the energies studied are dominated by simultaneous excitation or ionisation of the target. This
produces the energy deficit $\Delta E$ for these detached electrons, which we show is due to
the excitation of the target. Peak $P_0$, with its narrow angular spread, is related to
processes where the hydrogenic core $1s_\alpha$ is the one colliding with the target, while the
diffuse orbital is detached through a type of shake-off process. We show that excitation
of the target also prevails for this region of the electron distribution but only for light
targets such as He; for heavier atoms such as Ar we find that elastic dispersion of the
target produces an important contribution to detachment.

We also consider the contribution of double electron loss processes to the electron
distribution. When analysing DEL for one of the electrons emitted with velocity $v_e = v_i$, there appears the well known cusp $|v_e - v_i|^{-1}$ in the DDCS produced by the long-range
coulomb potential of the projectile nucleus (we neglect the screening by the other
electron, assumed to be ejected with a larger velocity in the projectile frame). The measured DDCS is the average of such a diverging distribution over the resolution
volume of the detector. This results in a peak for the DDCS whose height and width depend on the resolution, no matter how good this is. This feature can be used to separate electrons originating in SEL from DEL in the region $v_e \approx v_i$ by measuring spectra with different resolutions: the fraction of the distribution which remains constant in the limit of zero resolution volume should belong to SEL processes.

2. Single electron loss

We consider the usual formulation of high-energy electron detachment collisions (Lee
and Chen 1979, Franz et al 1981) to treat a transition where the target B goes from
the $\beta_i$ to the $\beta_f$ state. The bound state $\alpha_i$ of $\mathrm{H}^-$ will be approximated by a contracted
$1s_\alpha$ and a diffuse $1s_d$ orbital (Shull and Lowdin 1956), and the final state $\alpha_f$ represents
a free electron approximated by a plane wave $|k_e\rangle$, and a bound orbital with quantum
numbers $nl$ (atomic units are used throughout):

$$
|\alpha_i\rangle = N_i[|1s_\alpha(1)1s_d(2) + 1s_d(2)1s_\alpha(1)|], \\
|\alpha_f\rangle = N_f[|k_e, nl\rangle - \langle\alpha_i|k_e, nl|\alpha_f\rangle] 
$$

with

$$
1s_p(r) = (\gamma_p^3/\pi)^{1/2} \exp(-\gamma_p r), \quad \gamma_c = 1.04, \quad \gamma_d = 0.28
$$

$$
|k_e, nl\rangle = |k_e(1)nl(2) + k_e(2)nl(1)|
$$

The DDCS in the first-order Born approximation is

$$
\frac{d^2\sigma}{dv_e} = (2\pi)^{-4}v_e^{-1} \int dP \, P^{-4} \left| F_{\alpha_i,\alpha_f}(P) \right|^2 \left| F_{\beta_f,\beta_i}(P) \right|^2 \delta(P \cdot v_i - \Delta E) 
$$

with the kinetic energy loss

$$
\Delta E = E_{\beta_f} - E_{\beta_i} + E_{\alpha_f} - E_{\alpha_i} = \Delta E_B + k_e^2/2 + E(nl) - E(\mathrm{H}^-)
$$

where the electronic momentum $k_e$ is equal to the velocity $v_e'$ in the projectile frame: $k_e = v_e' = v_e - v_i$, and the generalised form factor for an atom with atomic number $Z$
and $N$ electrons for a momentum transfer $P$ is:

$$
F_{\delta_i,\delta_f}(P) = \left\langle \delta_f \left| Z - \sum_{j=1}^N \exp(iP \cdot r_j) \right| \delta_i \right\rangle.
$$
We are interested in separating the partial contributions of processes where the target is left in the initial state from those where it is excited or ionised, so we separate out the $\beta_j = \beta_i$ cross section in (2), and sum over the $\beta_i \neq \beta_j$ inelastic processes. The corresponding electron distributions are

$$n_1(E_e, \Theta_e) = \sum_{f \neq i} d^2\sigma / dE_e d\Omega_e = v_e \sum_{f \neq i} d^2\sigma / dv_e$$

(5)

$$n^*_1(E_e, \Theta_e) = d^2\sigma / dE_e d\Omega_e |\beta_j = \beta_i = v_e d^2\sigma / dv_e |\beta_j = \beta_i$$

(6)

where $E_e, \Theta_e$ are the energy and ejection angle of the electron in the laboratory frame.

The energy gained by the target is approximated by an average value, where the statistical weight is the probability that the target is excited to the $f$ state for a given momentum transfer $P$ (i.e. when the projectile is deflected along a given direction $\Theta = \Theta(P)$):

$$\Delta E_{\beta}(P) = \sum_{f \neq i} (E_{\beta} - E_{\beta_i}) d\sigma_{\beta f}(P) \left[ d\Omega \left( \sum_{f \neq i} d\sigma_{\beta f}(P)/d\Omega \right)^{-1} \right]^{-1}$$

(7)

(Lee and Chen 1979); the cross sections are evaluated in the first-order Born approximation, and the summations calculated by means of well known sum rules (Inokuti 1971)

$$\Delta E_{\beta}(P) = P^2 / 2S(P)$$

(8)

where

$$S(P) = \sum_{f \neq i} |F^0_{\beta f}(P)|^2$$

(9)

is the incoherent scattering function of atom B. The approximate $\Delta E_{\beta}$ used by Maleki and Macek is independent of $P$, and appreciably smaller than the values obtained from (8). Replacing the energy loss $\Delta E$ in the delta function of (2) by its average (closure approximation)

$$\overline{\Delta E}(P) = P^2 / 2S(P) + k_e^2/2 + E(nl) - E(H^-)$$

(10)

we obtain

$$n_1(v_e) = (2\pi)^4 v_e^{-1} \int dP P^{-4} |F^\Delta_{\alpha,\alpha}(P)|^2 S(P) \delta(P \cdot v_e - \overline{\Delta E}(P)).$$

(11)

The $v_e$ dependence of the electron distributions (5), (6) is introduced by the projectile form factor

$$F^\Delta_{\alpha,\alpha}(P) \sim \langle nl | 1s_d \rangle \langle k_e | \exp(-iP \cdot r) | 1s_d \rangle + \langle k_e | 1s_d \rangle \langle nl | \exp(-iP \cdot r) | 1s_d \rangle$

$$+ \langle nl | 1s_d \rangle \langle k_e | \exp(-iP \cdot r) | 1s_d \rangle + \langle k_e | 1s_d \rangle \langle nl | \exp(-iP \cdot r) | 1s_d \rangle$$

$$- F^\Delta_{\alpha,\alpha}(P) \langle \alpha | k_e, nl \rangle.$$
Electrons detached from fast $H^-$

The five terms of equation (12) that contribute to the $H^-$ form factor, evaluated for the minimum momentum transfer corresponding to each value of the electron ejection velocity $v_e$:

$$P_{\text{min}} = \frac{\Delta E_b + (v_e - v_i)^2/2 + E(1s) - E(H^-)}{v_i}$$

and considering the final H atom to remain in the ground $1s$ state.

width $\gamma_d$, so the transferred momentum $P$ leads to final states with a $v_e'$ distribution of similar width, given by $|v_e' + P| \sim \gamma_d$. In equation (11), energy conservation fixes the minimum value $P$ through the delta function, while the rest of the integrand is a decreasing function of $P$. Therefore, we expect the electron distribution arising from the first term of (12) to peak at $v_e' = P_{\text{min}} = \Delta E / v_i$. This is verified by the low-energy peak in the results presented in figure 1.

The second term in equation (12) describes the electron in the contracted orbital $1s_c$ making a transition to the $nl$ state with transferred momentum $P$: $\langle nl|\exp(-iP \cdot r)|1s_c\rangle$ and the diffuse electron, left in the field of the H($nl$) atom, preferentially populating low-energy continuum states. The $k_e$ dependence is given by the momentum transform $\langle k_e|1s_d\rangle$ of the diffuse orbital, which is concentrated around $v_e = v_n$ ($k_e = 0$), with a width $\delta k_e \sim \gamma_d$.

The third and fourth terms in equation (12) give small contributions compared with the ones presented because they involve projections of diffuse over contracted orbitals, and transitions peaked at large momentum transfers. Finally, the fifth term subtracts from the form factor those contributions coming from the non-orthogonality between initial and final states. It dominates the transition amplitude at the minimum momentum transfer considered in figure 1, making the amplitude negative at $v_e = v_i$. It enhances the presence of the two peaks by lowering the amplitude between them.

For the distribution $n^0_e(v_e)$, the momentum transfer $P$ should be small in order to leave the target unexcited. We expect those collisions to be soft with a large impact parameter. They will affect the diffuse orbital only, and therefore the momentum distribution of the ejected electron, equation (6), will be centred at $v_e = v_i$ with dispersion of order $\gamma_d$. 

Figure 1. The five terms of equation (12) that contribute to the $H^-$ form factor, evaluated for the minimum momentum transfer corresponding to each value of the electron ejection velocity $v_e$.
3. Double electron loss

We will extend our calculations to processes where one of the electrons of H\textsuperscript{-} is ionised to a state with negligible momentum $k_e$ in the frame of the projectile, while the other electron is ejected with momentum $K_e \gg k_e$. We assume $\langle r | k_e \rangle$ to be a continuum hydrogenic orbital normalised to be a plane-wave momentum eigenstate far away from the nucleus, so it behaves as $k_e^{-1/2}$ close to it (Salin 1969, Macek 1970, Ponce and Meckbach 1981).

The approximation in the description of the final state is similar to that made for SEL, but the bound state $nl$ is now replaced by a continuum state with small momentum $k_e$, neglecting the correlations and screening of the nuclear charge by the fast electron. We assume here that the slow electron screens the proton so that the fast electron can be described by a plane-wave state as in equation (1').

To obtain the resulting DDCS close to $\nu_e$, we integrate the double-electron distribution

\[
n_e^2(k_e, K_e) = \sum_{f \neq i} \frac{d^2 \sigma}{d E_e d \Omega_e} \, dK_e
\]

\[
n_e^2(k_e, K_e) = \frac{d^2 \sigma}{d E_e d \Omega_e} \, dK_e|_{\beta_f - \beta_i}
\]

over momentum $K_e$ of the fast electron. The result can be presented as:

\[
n_e^2(\nu_e) = f_c(2\pi/k_e)^2 F_e(\nu_e, k_e)
\]

where $\nu_e = \nu_i + k_e$, $F$ is only weakly dependent on $k_e$ and $f_c$ is the Coulomb factor which introduces the cusp on the distribution:

\[
f_c(x)^2 = \frac{1}{x} \quad \text{when } x \to 0.
\]

We have calculated $F$ separately for both elastic and inelastic dispersion of the target, and found a value of order 0.1 for 0.5 MeV collisions of H\textsuperscript{-} in He and Ar targets. Therefore, the contribution of DEL to the electron distribution will be negligible compared with that of SEL, except for $\nu_e = \nu_i$. Due to the cusp-like behaviour of the DEL contributions (14), (15) the measured distribution will depend on the resolution volume of the electron detector. We have performed the integration of equations (14), (15) over the mean resolution indicated by Menendez et al (1977) for their apparatus. Due to the diverging nature of the DDCS, the results for $\nu_e = \nu_i$ will be very dependent on the detailed form of the detector resolution function and the beam–target geometry.

4. Results

We present in figure 2 the contributions to the DDCS observed at an angle of 0.3° in the laboratory frame, arising from SEL and DEL processes for a He target, which is either left in the ground state or suffers an inelastic transition. The combined results are shown in figure 3 for several angles of ejection. The total SEL contribution is similar to that of Franz et al (1981), but the inclusion of DEL which enhances peak P\textsubscript{1} and the separate calculation of the various contributions as given in figure 2 allow us to have a better understanding of the intervening physical processes. The trailing peak P\textsubscript{1} is due to inelastic processes for the target, where the diffuse electron of H\textsuperscript{-} is dispersed by the target in much the same way as if it were a free electron with velocity...
Electrons detached from fast $H^-$

Figure 2. DDCS in 0.5 MeV $H^-$ collisions on He for $\theta_e = 0.3^\circ$, for the cases where there is single or double electron loss, with or without target excitation, defined by equations (5), (6), (13) and (14).

Figure 3. Total electron distribution at $\theta_e = 0.3$, 1.5 and 4$^\circ$, for 0.5 MeV $H^-$ collisions on He (full curves). Experimental results (Menendez and Duncan 1979) are shown as broken curves, and are normalised to the calculations at the maximum of peak $P_1$ for $\theta_e = 0.3^\circ$. This peak is clearly defined in the measurements, and it should not be affected by uncertainties in the resolution of the electron detector, as may be the case of peak $P_0$.

distribution $v_e = v_i + k_e$. The contracted electron of $H^-$ is simply a spectator in this first-order Born approximation. The average excitation energy of the He target in the closure approximation, equation (8), is of the order of 40 eV for the momentum transfers $P$ relevant to collisions with 0.5 MeV $H^-$ ions; therefore, ionisation of the target should be the dominant process. Figure 2 verifies that the trailing peak $P_1$ is centred at

$$v_e = v_i - \frac{\Delta E(P)}{v_i} \bigg|_{P=v'_i}$$

and that it has a relatively wide angular spread, as it corresponds to free electrons scattered by an atom. The peak centred at $v_e = v_i$ is mainly due to inelastic collisions of the core $1s_e$ state of $H^-$ with the He target while the weakly bound electron acts as a spectator, and is finally detached. Figure 2 shows that the events where the target is left in the initial state are only a small fraction of the total.

Figure 4 shows the separate contributions of $SEL$ and $DEL$, with and without target excitation, to the DDCS at 0.8$^\circ$ in the laboratory frame, from 0.5 MeV $H^-$ collisions on Ar. The results are similar to those for He, but now processes where the target remains in its ground state start to be important both for $SEL$ and $DEL$. This is due to the presence of tightly bound electrons in the target which scatter off the $H^-$ electrons without gaining the energy needed to become excited.
Figures 3 and 5 show the overall accord of the velocity and angle dependence of the calculated electron distribution with measurements. They also show some discrepancies, such as the valley between peaks, which is deeper, and the peaks which are narrower than in measurements. This may be due to the closure approximation employed, which amounts to the replacement of the energies gained by the target by a unique average value, which therefore concentrates the trailing peak $P_1$ making it narrower. This effect may also have some influence on the shift of the calculated peak $P_1$ for Ar targets, towards lower energies compared with measurements shown in figure 5.

5. Conclusions

We have calculated the distributions of electrons ejected from $H^-$ by collisions with He and Ar atoms.

Single-electron loss processes produce two peaks on the energy distribution of the electrons, which correspond to binary collisions of either the weakly or the tightly bound electrons of $H^-$ with the target. The closure approximation, used to simplify the sum over final states of the target, gives $\Delta E(P_{\text{min}})/v$, as the velocity separation between the peaks, where $\Delta E(P_{\text{min}})$ is the average excitation energy of the target for...
Electrons detached from fast $H^-$

the minimum momentum transfer $P_{\text{min}}$. Processes where the target scatters elastically, that were known to give negligible contribution for He targets, produce an appreciable number of electrons with velocities close to that of the projectile for Ar targets. This should also be the case for other multielectron targets.

We have shown that double electron loss of $H^-$ contributes appreciably to the measured DDCS, therefore the experimental results should depend on the detector resolution as happens with measured distributions of electrons on continuum states of Coulomb potentials.

The wavefunctions used are simple products of 1s orbitals for the initial state, a hydrogen 1s orbital plus a plane wave for single electron loss, and a Coulomb wave plus a plane wave for double ionisation. This neglects correlations between electrons in the final state, which may be justified because in the Born approximation ionisation is produced by a binary collision between the electron and the target atom, and the diffuse orbital which acts as initial state has a maximum in the probability density at large distances from the residual H atom. At those distances it may be reasonable to neglect electron correlations and consider total screening of the proton charge by the inner electron. Further work is needed to assess the importance of electron correlations in the initial and final states, and more so for the case of double electron loss, where states with two continuum electrons of similar energy may be populated, and could lead through angle correlations to anisotropies in the resulting electron distribution.

The closure approximation, which replaces the multitude of final target energies by an average value $\overline{\Delta E}(P)$, may be responsible for calculated peaks narrower than those observed, and for the shift of the trailing peak towards lower energies for Ar targets. Calculations of transitions to those definite final states of the target which are populated preferentially may correct these features.

Finally, the results for double electron loss are quite sensitive to the resolution volume of the electron detector; therefore, it will be of great interest to perform measurements of the electron distribution with variable resolution and in this way to separate the contributions of double and single electron loss.

References

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