

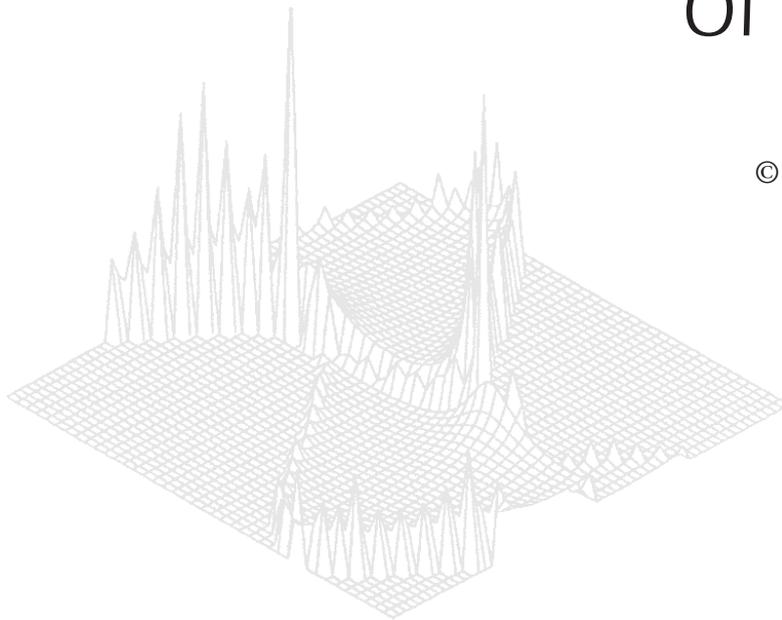
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## Coherence and Correlation in Electron Scattering from the Alkalis\*

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### *Abstract*

A series of superelastic electron scattering experiments from lithium and from potassium is described in which the total polarisation parameter  $P^+$  is measured. We report significant departures from the coherence condition  $P^+ = 1$  for both targets. The structure observed in the parameter  $P^+$  can be interpreted by a qualitative wave mechanical model that had been introduced by our research group to explain similar structure in superelastic electron scattering experiments from sodium.

### 1. Introduction

The results from superelastic electron scattering experiments have been shown to provide a very sensitive test of the theory of electron scattering from atoms. The technique involves scattering electrons from laser excited atoms and detecting those electrons that gain energy in the collision. A detailed picture of the final state wave function of the scattering process can be determined when the number of superelastically scattered electrons is measured as a function of the polarisation of the laser beam. The scattering parameters deduced from the experiments can be compared directly with those predicted from the theory. Our work on superelastic scattering from the alkalis, e.g. from sodium (Scholten *et al.* 1993), lithium (Karaganov *et al.* 1996) and potassium (Stockman *et al.* 1998) has, amongst other things, shown that the convergent close coupling approximation of Bray (1994) provides an exceptionally accurate description of the excitation of the first excited states of each of these alkali targets.

These experiments have also demonstrated that there is significant structure in the total polarisation parameter  $P^+$  (equation 3) at certain incident energies and that this structure has consistent properties over the range of alkalis studied. Teubner and Scholten (1992) first noted this structure in their superelastic scattering experiments from sodium. The general features of the structure were manifested by a significant departure from the condition  $P^+ = 1$  at low incident energies. The structure was more pronounced as the incident energy was reduced and it moved to larger angles as the energy was reduced. The minimum in  $P^+$

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corresponded to minima in the differential cross sections and also to zeros in the orientation parameter. Teubner and Scholten (1992) proposed a simple wave mechanical model to explain the general features of the observed structure. This model can clearly be applied to other alkali targets and indeed can be used to provide a qualitative description of the process in lithium and in potassium.

The basis of the phenomenon is the exchange scattering amplitude and correlation between the incident and bound electrons in the scattering process. Consequently it is appropriate that it be discussed in this special Australian–German workshop issue on Electron Correlations.

In this paper the experimental technique is briefly described in Section 2, we introduce appropriate theory in Section 3 and the results are presented and discussed in Section 4.

## 2. Experimental Technique

A schematic diagram of the experimental geometry is shown in Fig. 1. The apparatus and techniques used in the present work are described fully in Karaganov *et al.* (1998). Briefly, a collimated beam of alkali atoms was produced in a resistively heated oven. The atoms were optically pumped to the appropriate  $^2P_{3/2}$  state using laser radiation from a stabilised ring dye laser (Spectra Physics 380D). The collimated laser beam was incident normal to the scattering plane. For each of the alkali atoms involved in the present study the major limitation to optical pumping arose from the hyperfine structure trapping effect. These limitations were particularly severe and so it was necessary to use two frequency pumping from both ground state hyperfine structure levels. The way in which this was achieved is described by Karaganov *et al.* (1999). A well collimated electron beam of the required energy intersected the alkali and laser beams at the

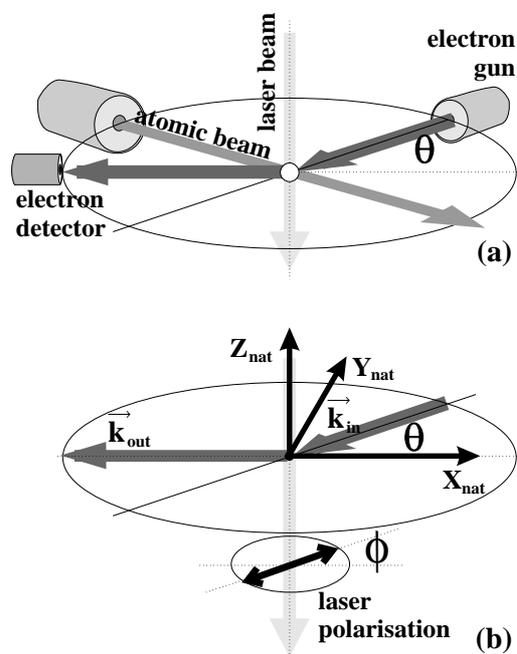


Fig. 1. (a) Schematic diagram of the superelastic electron–atom scattering apparatus. (b) Scattering geometry shown in the natural frame of reference (Hermann and Hertel 1982a, 1982b).

interaction region. The superelastically scattered electrons that had gained energy equivalent to that of the laser photons used to excite the atoms were analysed with an electrostatic energy analyser that viewed the interaction region. The electrons were detected by a channel electron multiplier. The electron scattering angle was changed by rotating the electron gun about the laser beam axis. The natural frame  $x$  axis, as defined by Herman and Hertel (1982*a*, 1982*b*), lies in the collision plane (Fig. 1). It was fixed parallel to the fast electron trajectory. This axis was the reference axis for the linear components of the laser polarisation.

The superelastic electron signal was monitored as a function of the polarisation of the laser beam and the three components of the equivalent Stokes vector were calculated from the relationships:

$$\begin{aligned} P_1 &= \frac{\mathcal{I}_0 - \mathcal{I}_{90}}{\mathcal{I}_0 + \mathcal{I}_{90}}, \\ P_2 &= \frac{\mathcal{I}_{45} - \mathcal{I}_{135}}{\mathcal{I}_{45} + \mathcal{I}_{135}}, \\ P_3 &= \frac{\mathcal{I}_{\text{RHC}} - \mathcal{I}_{\text{LHC}}}{\mathcal{I}_{\text{RHC}} + \mathcal{I}_{\text{LHC}}}. \end{aligned} \quad (1)$$

Here  $\mathcal{I}_\phi$  is the superelastic count rate when the laser light is polarised at an angle  $\phi$  with respect to the  $x$  axis, while  $\mathcal{I}_{\text{RHC}}$  and  $\mathcal{I}_{\text{LHC}}$  are the respective count rates when right-hand circularly and left-hand circularly polarised light are used for the pumping process.

The measured parameters defined in equation (1) were influenced by the incomplete polarisations achievable in the optical pumping process. In any event the fine and hyperfine structure relaxation times are so long compared to the collision time that the laser induced alignment and orientation can be factored out to yield the reduced Stokes parameters  $\bar{P}_i$ , as shown by Farrell *et al.* (1991) for the case of sodium:

$$\bar{P}_1 = \frac{1}{K} P_1, \quad \bar{P}_2 = \frac{1}{K} P_2, \quad \bar{P}_3 = \frac{1}{K'} P_3. \quad (2)$$

A similar approach was used in the present work. The superelastic depolarisation factor for linear pumping  $K$  was taken to be identical to the line polarisation  $P_L$  of the resonance fluorescence from the alkali beam (Teubner *et al.* 1996). The fluorescence signal was monitored through a window in the vacuum chamber during the experiment and it was therefore possible to determine  $K = P_L$  for each measurement in a similar manner to that suggested by Meng *et al.* (1992).

The factor  $K'$  that characterises the depolarisation in the case of circularly polarised optical pumping plays a very important role in the determination of  $\bar{P}_3$ . It is commonly accepted to assume that  $K' = 1$  because it can be shown (Farrell *et al.* 1991) that it is very close to unity at the laser intensities used in the present experiments. Indeed in their experiments on sodium, Scholten *et al.* (1993) measured  $K'$  and confirmed that  $K' = 1$ , within the experimental errors which were about 10%. The size of the relative error reflected the difficulty of the measurement.

From general considerations one can write

$$\frac{P_3}{K'} = \bar{P}_3 \leq 1 \quad \text{and} \quad K' \leq 1.$$

In the case of our experiments on lithium the maximum measured value of  $P_3$  was

$$P_3 = 0.986 \pm 0.033$$

and was observed at  $80^\circ$  for an electron energy of 20 eV. Thus, in the case of lithium the meaningful range of values of  $K'$  was

$$1 \geq K' \geq 0.986 \pm 0.033.$$

Similar conclusions could be drawn from our work on potassium.

The total reduced polarisation  $P^+$  is then defined from

$$P^+ = \sqrt{\bar{P}_1^2 + \bar{P}_2^2 + \bar{P}_3^2}. \quad (3)$$

### 3. Theoretical Considerations

The final state wavefunction for the excitation of  $^2P$  states in the alkali atoms can be written as a superposition of substates of  $|M_L\rangle$  basis states in the natural frame (Andersen *et al.* 1988):

$$|^2P\rangle = a_1|+1\rangle + a_{-1}|-1\rangle,$$

where the  $a_m$  are the scattering amplitudes.

Considering the two electrons involved in the collision, the spins can couple to yield either singlet or triplet wavefunctions. If unpolarised electrons are used, as is the case in the present experiments, the final state is described by an incoherent sum of the two wavefunctions that is appropriately weighted.

Andersen *et al.* (1988) have shown that in the case of superelastic scattering experiments performed with unpolarised electrons, the total polarisation  $P^+$  is related to the separate spin amplitude channels  $a_m$  by

$$(P^+)^2 = 1 - \frac{3}{4} \left| a_1^{(t)} a_{-1}^{(s)} - a_{-1}^{(t)} a_1^{(s)} \right|^2, \quad (4)$$

where the superscripts  $(t)$  and  $(s)$  represent the triplet and singlet amplitudes respectively. Here  $P^+$  directly reflects the total polarisation of the excited  $^2P$  state. For a pure triplet or singlet channel, the excitation of the separate  $|+1\rangle$  and  $|-1\rangle$  orbitals should be coherent with a fixed phase between the two amplitudes  $a_1$  and  $a_{-1}$ . Consequently we have  $P^+ = 1$ . However, with incoherently mixed spin states, equation (4) shows that  $P^+$  may be less than 1. Such departures from unity reflect the indeterminate phase relationship between the amplitudes. It follows from the loss of information that arises from the fact that spin is ignored in the present experiments.

Equation (4) can be rewritten following Kessler (1985) by converting the singlet and triplet amplitudes into direct (D) and exchange (E) amplitudes. In this case we get

$$(P^+)^2 = 1 - 3 |a_1^E a_{-1}^D - a_{-1}^E a_1^D|^2. \quad (5)$$

Equation (5) clearly shows that the condition  $P^+ < 1$  implies that the exchange amplitudes  $a_{\pm 1}^E$  are non zero. Consequently, a study of the total polarisation can provide information on the role of exchange in the collision process.

Teubner and Scholten (1992) showed that in the case of electron scattering from sodium, there were significant departures from the condition  $P^+ = 1$  at about the same scattering angles where there were minima in the differential cross sections. In that case they relied on previous measurements of the differential cross sections of Teubner *et al.* (1986). In the present work we have been able to measure the differential cross sections for the excitation of the  $^2P$  states at the same time as the Stokes parameters are measured. Here we follow the prescription of Andersen *et al.* (1988) who showed that in the natural frame the angular part of the final state wavefunction can be written as

$$\Psi \propto a_\sigma \cos \phi + a_\pi \sin \phi, \quad (6)$$

where  $a_\sigma$  and  $a_\pi$  are the scattering amplitudes in the atomic natural frame and  $\phi$  is the azimuthal angle with the  $x$  axis. There are simple relationships between these amplitudes and the  $a_{\pm 1}$ .

The angular part of the electron density then is

$$|\Psi|^2 \propto |a_\sigma|^2 \cos^2 \phi + |a_\pi|^2 \sin^2 \phi + 2\text{Re}(a_\sigma a_\pi^*) \sin \phi \cos \phi. \quad (7)$$

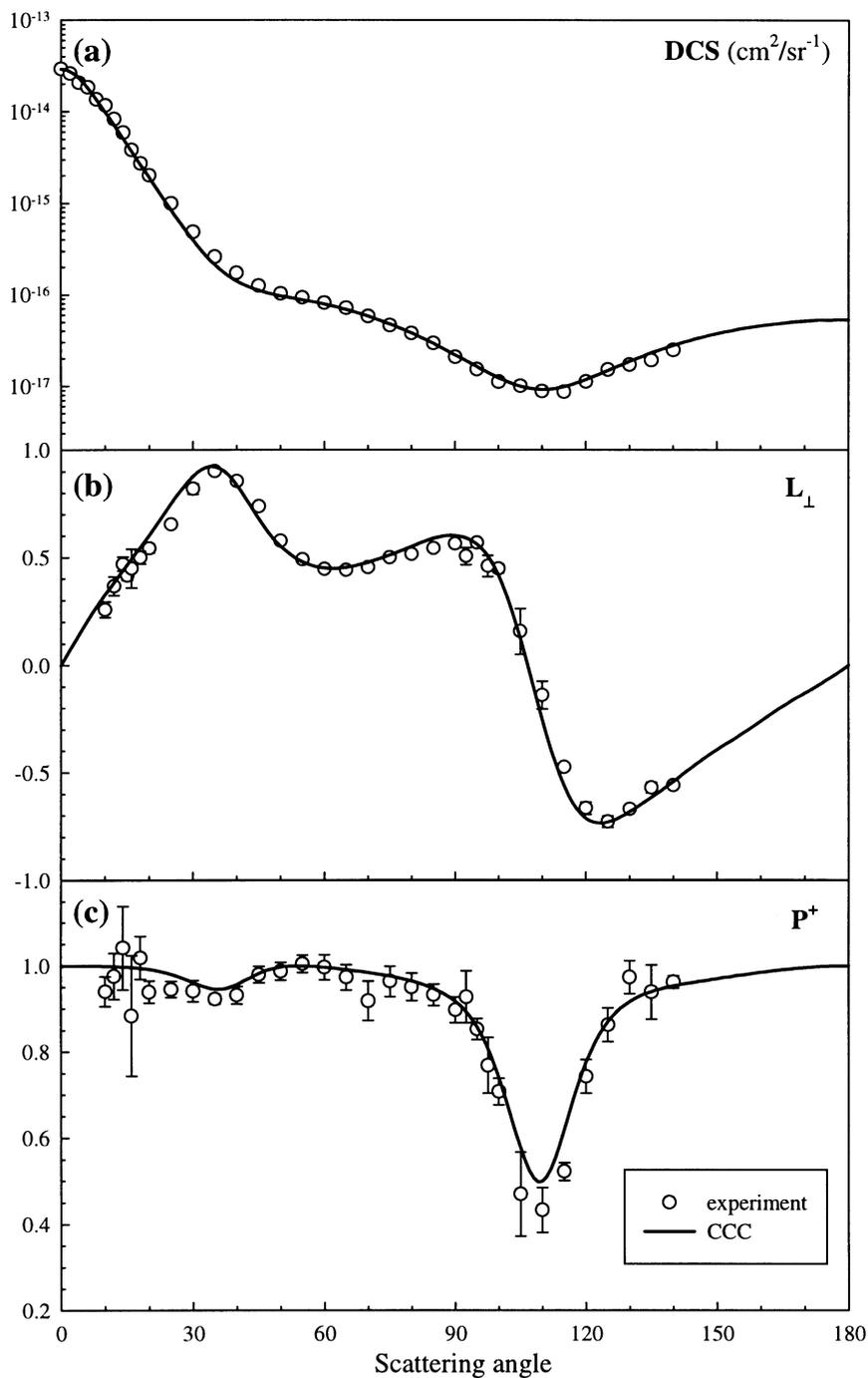
If one assumes that the charge cloud consists of two linear oscillators with amplitudes  $a_\sigma$  and  $a_\pi$  the expression for the sum  $I(0) + I(90)$  becomes

$$I(0) + I(90) \propto |a_\sigma|^2 + |a_\pi|^2 \propto \sigma(\theta), \quad (8)$$

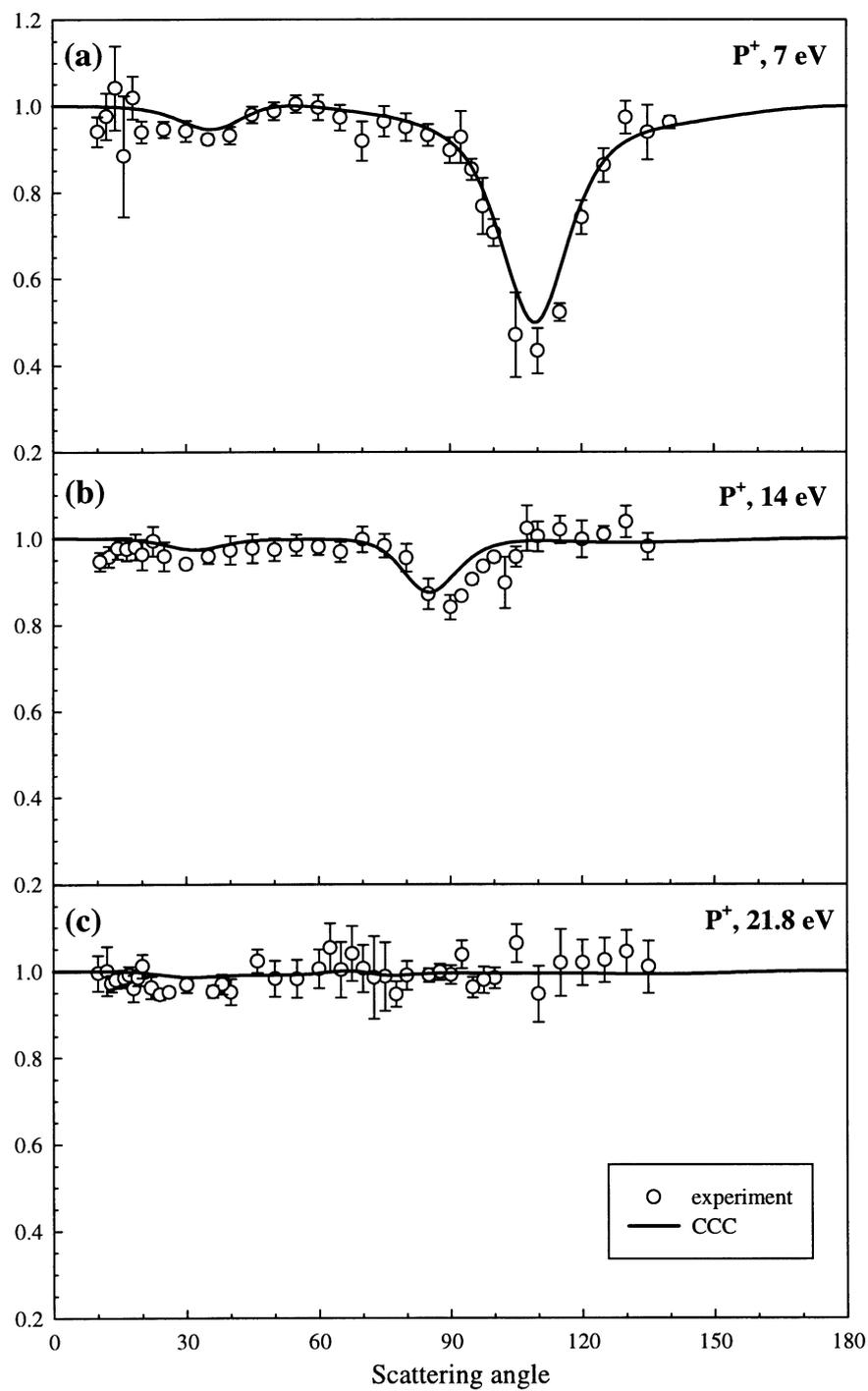
where  $\sigma(\theta)$  is the differential cross section.

#### 4. Results and Discussion

Fig. 2a shows the differential cross section for the excitation of the  $^2P$  state in lithium for an incident electron energy of 7 eV referred to the ground state. The cross section has been normalised by integrating the angular distribution given by equation (8) and normalising that integral to an interpolation of the total cross section measured by Leep and Gallagher (1974). We assume that the angular distribution for the  $^2P_{1/2}$  state has the same shape as that for the  $^2P_{3/2}$  state. The experimental results are compared to those predicted by the convergent close coupling calculation (Karaganov *et al.* 1999). There is excellent agreement between the two sets of data. The minimum in the differential cross section occurs at  $110^\circ$ .



**Fig. 2.** Measurements and convergent close-coupling (CCC) calculations of the differential cross section, orientation parameter  $L_{\perp} = \bar{P}_3$  and coherence parameter  $P^{+}$  for the 2S-2P transition in lithium at an electron energy of 7 eV referred to the ground state (Karaganov *et al.* 1998).



**Fig. 3.** Measurements and convergent close-coupling (CCC) calculations of the coherence parameter  $P^+$  for the 2S-2P transition in lithium at electron energies of 7 eV, 14 eV and 21.8 eV referred to the ground state (Karaganov *et al.* 1998).

Fig. 2c shows the total reduced polarisation  $P^+$  at an incident energy of 7 eV referred to the ground state. The dominant feature of  $P^+$  is a very deep minimum at  $110^\circ$  which closely matches the position of the minimum in the differential cross section. This is consistent with the observations of  $P^+$  in sodium made by Teubner and Scholten (1992) whose qualitative explanation of the phenomenon can also be used in the present case. At 7 eV the de Broglie wavelength of the continuum electron is comparable to the dimensions of the atom. Thus, the angular distribution in Fig. 2a can be considered as the Fraunhofer diffraction pattern of the atom. Hence, there will be a minimum in the pattern at an angle  $\theta$  where  $\sin\theta \simeq \lambda/D$  and  $D$  is the atomic dimension. Electrons scattered at this angle are such that there is maximum overlap between the wavefunctions for the continuum electron and that of the bound electron. The exchange amplitudes, which arise from the overlap between the wavefunctions of the two electrons will be a maximum at this angle. Consequently structure will be observed in  $P^+$ .

In Fig. 2b we show the parameter  $\bar{P}_3$  where

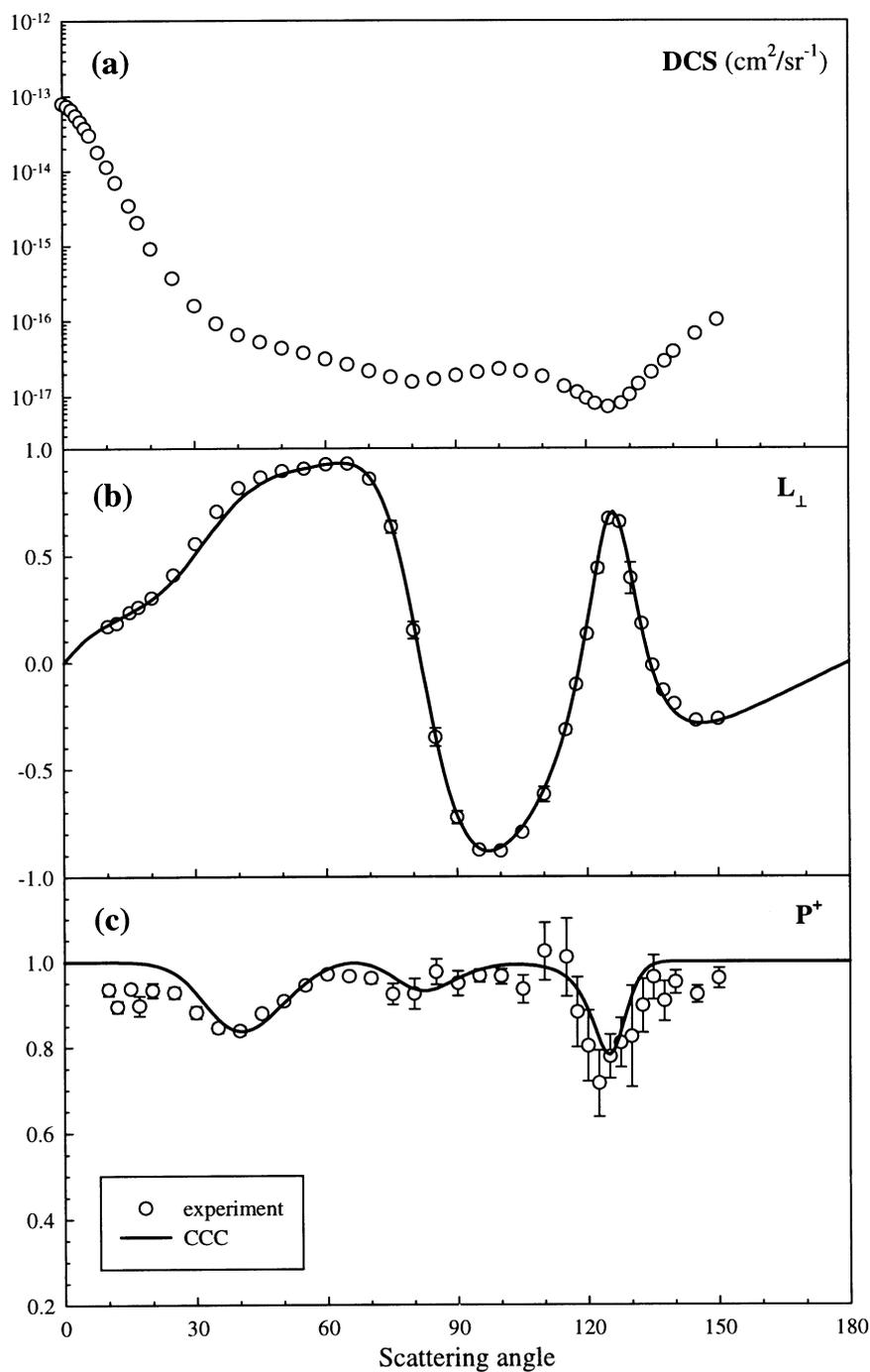
$$\bar{P}_3 = -L_\perp \quad (\text{Andersen } et al. 1988)$$

and  $L_\perp$  is the angular momentum transferred to the atom normal to the scattering plane. It is clear that  $L_\perp$  is zero at about the same scattering angle that  $P^+$  is a minimum. This fact cannot be explained on arithmetic grounds because  $P^+$  arises from the addition of three terms in quadrature. The results are, however, consistent with the hypothesis that the angular momentum is transferred to the sodium atom through the atomic electron. Consequently, similar conditions apply for both maximum exchange and for zero angular momentum transfer.

As the incident energy increases one would expect the exchange amplitudes to decrease. The de Broglie wavelength  $\lambda$  will decrease so the angle at which the minimum in  $P^+$  is seen will also decrease. Fig. 3 shows  $P^+$  for energies of 7 eV, 14 eV and 21.8 eV relative to the ground state. The minimum at 14 eV is at  $90^\circ$  which is clearly less than that at 7 eV. The depth of the minimum at 14 eV is also less than that at 7 eV reflecting the decrease in the exchange amplitudes. No minimum is observed in  $P^+$  at 21.8 eV despite the observed minimum for sodium observed by Teubner and Scholten (1992) at about this energy.

Fig. 4 summarises our results for superelastic scattering from the  $4^2P$  state in potassium at an energy of 10 eV referred to the ground state. The differential cross section in Fig. 4a shows considerably more structure than was the case for lithium and reflects the increase in size of the potassium atom. In this case the angular distribution was integrated using Simpson's rule and normalised to an interpolated integral cross section for the excitation of the  $4^2P$  state of Chen and Gallagher (1978).

The structure in  $P^+$ , shown in Fig. 4c, matches both minima in the angular distribution curve. There is however an additional minimum at around  $40^\circ$  that cannot be explained using our simple model. Technically it arises because  $\bar{P}_1$  and  $\bar{P}_2$  are both nearly zero here. It does however, correspond to a point of inflection in the angular distribution. Nevertheless, the minima in the differential cross sections at  $80^\circ$  and  $120^\circ$  reflect the structure in  $P^+$  at these angles. There is a zero in the angular momentum transfer at about  $80^\circ$  but the other zero in the angular momentum transfer does not occur at  $120^\circ$ .



**Fig. 4.** Measurements and convergent close-coupling (CCC) calculations of the differential cross section, orientation parameter  $L_{\perp} = \bar{P}_3$  and coherence parameter  $P^{+}$  for the 4S-4P transition in potassium for at an electron energy of 10 eV referred to the ground state (Stockman *et al.* 1998).

## 5. Conclusions

The present study of the total polarisation parameter  $P^+$  in superelastic electron scattering experiments in lithium shows that at low incident energies there are significant departures from the condition  $P^+ = 1$  at middle scattering angles. This structure becomes less pronounced and moves to smaller scattering angles as the energy increases. At an energy of about ten times the excitation energy no structure is observed. This structure is consistent with that observed in sodium by Teubner and Scholten (1992) who gave a qualitative explanation based on the relative importance of the exchange scattering amplitudes. Similar structure is observed in our superelastic scattering experiments on potassium, but in this case the complexity of the target structure demonstrates that the simple model cannot explain all of the structure observed in  $P^+$ .

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