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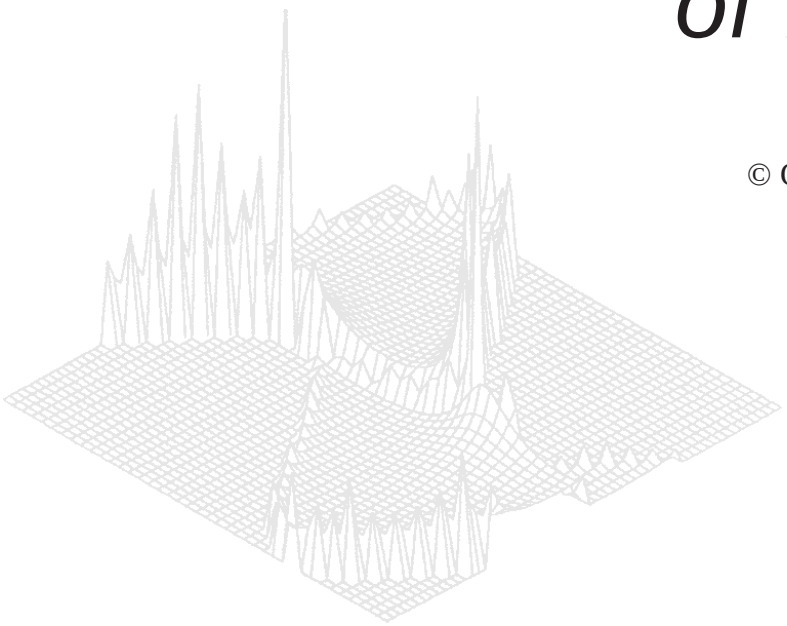
Australian Journal of Physics

Volume 52, 1999



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Aust. J. Phys., 1999, **52**, 515–22

Transfer of Angular Momentum in Electron Collisions with Alkali Atoms*

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Abstract

Measurements of the transfer of angular momentum to rubidium and sodium atoms in collisions with electrons are reported. For excitation of the rubidium $5^2S_{1/2}$–$5^2P_{3/2}$ transition, it is found that existing first order distorted wave Born approximation calculations show poor agreement with the data and that a model which includes the relativistic interaction between the electrons and the atoms in the potential is needed. For the de-excitation of the sodium $4^2S_{1/2}$–$3^2P_{3/2}$ transition, a long standing proposal relating to the sign of the transferred angular momentum is not supported except at small scattering angles. A convergent close coupling calculation displays excellent agreement with the measured data.

1. Introduction

Electron collisions with atoms can be characterised by a number of physical properties. While cross sections provide information concerning the probability of an interaction occurring, it is the measurement of electron–atom correlations and coherences that enables detailed information about scattering amplitudes to be obtained. A useful parametrisation of the scattering amplitudes is obtained in the Natural Frame (Andersen *et al.* 1988) in which atomic collision parameters (ACPs) have been defined to enable the alignment and orientation of the atomic charge cloud following the collision to be determined. For electron excitation of an S state to a P state, up to four ACPs can be measured which completely describe the collision process (except for identifying the spin of the electrons).

Of particular interest is the transfer of angular momentum to the atom during the collision. This transfer occurs perpendicular to the scattering plane defined by the incident and scattered electron directions and is commonly given the notation $L_\perp$. This parameter has evoked considerable theoretical interest particularly when attempting to interpret the results in terms of classical trajectories and the sign of $L_\perp$ to the attractive or repulsive nature of the interaction potential (Kohmoto and Fano 1981; Hermann and Hertel 1980). It is also worth noting that the simplest theoretical description of an electron–atom collision, the first Born approximation (FBA), does not predict any transfer of angular momentum in the collision process.

* Refereed paper based on a contribution to the Australia–Germany Workshop on Electron Correlations held in Fremantle, Western Australia, on 1–6 October 1998.

 10.1071/PH98097 0004-9506/99/030515$05.00

Although the sign of $L_\perp$ cannot be *a priori* predicted, its variation with scattering angle θ seems to follow a pattern for electron excitation to the first P state in light atoms such as hydrogen and helium and the alkalis. For reasons of axial symmetry, $L_\perp$ must be zero at $\theta = 0°$. As θ increases from 0°, $L_\perp$ has increasing positive values until it reaches a maximum, decreases until changing sign, passes through a minimum before again tending to zero at 180°. Maxima and minima values close to unity in magnitude mean that the atom is almost completely oriented following the collision. The alkali atoms investigated to date have shown additional structure. For lithium at incident electron energies of 7 and 14 eV (Karaganov *et al.* 1998) and sodium at 22·1 eV (Scholten *et al.* 1993), $L_\perp$ passes through an additional small minimum and maximum of positive values before changing sign. For the heavier alkali, potassium, at 10 eV (Stockman *et al.* 1998), the value of $L_\perp$ has an extra maximum and minimum together with two more changes in sign at larger scattering angles following the first minimum.

Information on the importance of terms in a Born series expansion of the scattering amplitudes has been discussed in relation to the shape of $L_\perp$ (Madison and Winters 1981; Andersen and Hertel 1986). While the first non-zero term dominates at lower scattering angles, the change in sign of $L_\perp$ indicates that the next highest order term, which has a different sign, has become greater in magnitude. The first term, which is the product of a first- and second-order amplitude, is proportional to the cube of the charge and so, it was argued, that $L_\perp$ would be negative for positron excitation at small scattering angles. It was further argued that, as the first term changed sign under reversal of direction of energy transfer, de-excitation of an S state to a P state would also yield negative values of $L_\perp$.

In this work, reported at the Australia–German Workshop on Electron Correlations in October 1998, we present two studies of the transfer of angular momentum in electron–alkali atom collisions. Firstly, we have obtained measurements of $L_\perp$ for excitation of the $5^2S_{1/2}$–$5^2P_{3/2}$ transition of Rb, the heaviest alkali studied to date. Secondly, we have tested the hypothesis predicting a change in sign of $L_\perp$ with change in energy tranfer direction, by measuring the parameter for the de-excitation process $4^2S_{1/2}$–$3^2P_{3/2}$ for Na.

2. Experiment

We employ the technique in which measurements of $L_\perp$ are made by colliding electrons with the P state which has been excited and optically prepared by circularly polarised laser radiation. A schematic of this arrangement is shown in Fig. 1. Laser radiation is injected into the interaction region perpendicular to the scattering plane defined by the momenta of the incident and scattered electrons. For rubidium, those electrons which have de-excited the atoms from the $5^2P_{3/2}$ level to the $5^2S_{1/2}$ ground level and gained the transition energy of 1·6 eV are detected after passing through a cylindrical electron analyser. These electrons are said to have been *superelastically scattered* (Hertel and Stoll 1974). The experiment is run in the time-reverse mode to the conventional coincidence experiment in which the electron excites the transition and the subsequently emitted photon is detected in coincidence with the inelastically scattered electron. Data collection rates for superelastic scattering experiments are many orders of magnitude faster than for coincidence experiments since only one product of the collision, the scattered electron, needs to be detected.

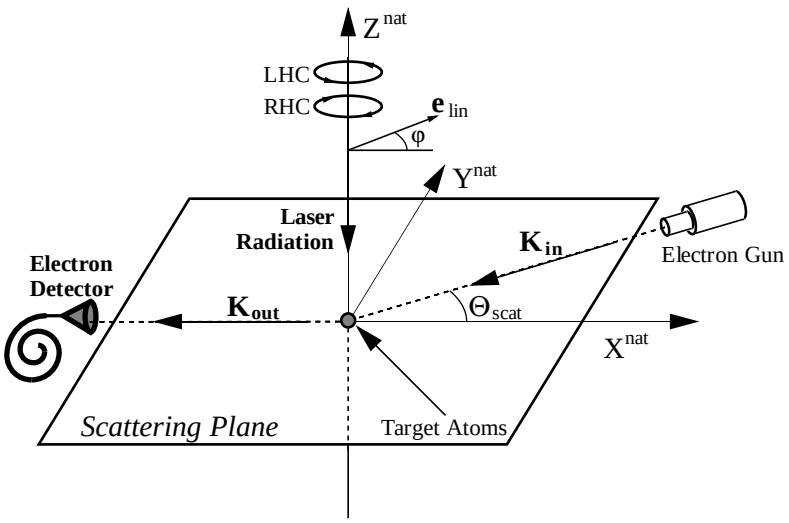

Fig. 1. Geometry of the experiments performed in time-reversed mode. The axes for the natural frame are shown.

It is possible to relate the superelastic parameters to those defined for direct electron excitation of the transition. To extract values of $L_\perp$, the pseudo Stokes parameter P_3^{S} is measured (Farrell *et al.* 1991):

$$P_3^{\mathrm{S}} = \frac{S_{\mathrm{RHC}}(\theta) - S_{\mathrm{LHC}}(\theta)}{S_{\mathrm{RHC}}(\theta) + S_{\mathrm{LHC}}(\theta)} = -K' L_\perp \,, \tag{1}$$

where $S_{\mathrm{RHC}}(\theta)[S_{\mathrm{LHC}}(\theta)]$ is the count rate of electrons scattered at angle θ from the target atoms excited by right-hand [left-hand] circularly polarised laser radiation. Here K' describes the optical pumping within the hyperfine substates of the $5^2\mathrm{P}_{3/2}$ level and is calculated using a full quantum electrodynamical model for the laser excitation of the transition.

To determine $L_\perp$ for the $4^2\mathrm{S}_{1/2}$–$3^2\mathrm{P}_{3/2}$ Na transition, the same time-inverse experiment can be utilised. In this procedure, however, those electrons that are *inelastically scattered* from the P level and excite the atoms to the $4^2\mathrm{S}_{1/2}$ level are detected. Using the time-reverse geometry, the transfer of angular momentum for the de-excitation process can be deduced from equation (1).

For an atom that spends many lifetimes passing through the laser beam, circularly polarised excitation results in optical pumping to a two state atomic system. For example, excitation of the ^{85}Rb $5^2\mathrm{S}_{1/2}$–$5^2\mathrm{P}_{3/2}$ transition using left-hand circularly polarised radiation (σ^+ excitation), will lead to the atom being constrained to cycle between the $5^2\mathrm{S}_{1/2}(F = 3,\ m_{\mathrm{F}} = 3)$–$5^2\mathrm{P}_{3/2}(F = 4,\ m_{\mathrm{F}} = 4)$ substates as long as the atom remains in a resonant condition. As these states are pure states in L-representation, there should be no need to compensate for a distribution within the hyperfine substate manifold and K' should have a magnitude of unity. In practice, complete optical pumping to these pure L-states is not achieved in the ensemble of atoms averaged over its time distribution in the laser beam. For the ^{85}Rb $5^2\mathrm{S}_{1/2}$–$5^2\mathrm{P}_{3/2}$ transition, K'

takes the value of $-0{\cdot}985$ for the conditions of our experiment. The value of K' is $-0{\cdot}982$ for the same transition in the ^{87}Rb isotope while, for excitation of the $3^2S_{1/2}$–$3^2P_{3/2}$ Na transition, it is $-0{\cdot}99$. In the latter case, radiation from two lasers excite Na atoms from both hyperfine states of the ground level. This enables the excited level population to be maximised, which is necessary because of the small signal-to-noise ratio when detecting the scattered electron for this excited state to excited state transition.

The apparatus for these experiments has been described previously (Sang *et al.* 1994; Shurgalin *et al.* 1998) and so only a brief summary will be given here. For the rubidium experiment, a non-energy selected gun incorporating a BaO filament in conjunction with an electron detector consisting of a cylindrical analyser and channeltron produced a resolution of 300 meV at 20 eV electron energy. The sodium experiment required a system with higher resolution. This was achieved using a gun consisting of a tungsten filament and a hemispherical selector together with a hemispherical analyser in the detection system. This electron spectrometer had a resolution of 150 meV at 20 eV electron energy.

The atomic ovens were standard heated crucible types consisting of a reservoir and differentially heated nozzle. The rubidium oven produced a beam of ^{85}Rb and ^{87}Rb in their natural abundance (72%:28%). A Doppler width of 200 MHz was estimated from resonance fluoresence measurements. The sodium atomic beam was further collimated by inserting cooled apertures yielding a residual Doppler width of 100 MHz.

The electron spectrometers and atomic ovens were housed in stainless steel chambers evacuated by turbo molecular pumps. The base pressure in each chamber was approximately 10^{-7} mbar. The magnetic field in the interaction regions was reduced to below $0{\cdot}8$ mT in any direction by internally lining the chambers with 2 mm thick m-metal and using three orthogonal pairs of Helmholtz coils externally.

Laser radiation was provided by tunable lasers pumped by an Ar^+ laser. In the Rb case, a titanium : sapphire laser was tuned to $780{\cdot}24$ nm to excite the $5^2S_{1/2}$–$5^2P_{3/2}$ transition, while for Na, dye lasers operating at $589{\cdot}16$ nm using rhodamine 590 were tuned to excite the atoms from the F = 2 and F = 1 hyperfine ground states respectively. Before entering the vacuum chambers, linearly polarised radiation from the lasers passed through quarter wave retarders to generate circularly polarised light of the required handedness. This handedness was reversed by rotation of the retarder through 90° by a stepper motor controlled by a personal computer.

For both experiments, the output pulses from the channeltrons were amplified and counted using standard NIM electronics. Counts were recorded on the personal computer for a given time at each handedness of the polarisation. Background counts were also recorded by blocking the laser beam. The value for the pseudo-Stokes parameter was calculated by the computer for each run which was repeated at least twenty times and then averaged.

3. Results

Measured values of $L_\perp$ as a function of scattering angle θ for excitation by incident electrons of 20 eV energy of the $5^2S_{1/2}$–$5^2P_{3/2}$ transition of rubidium are presented in Fig. 2. This is equivalent to an incident electron energy of $18{\cdot}4$

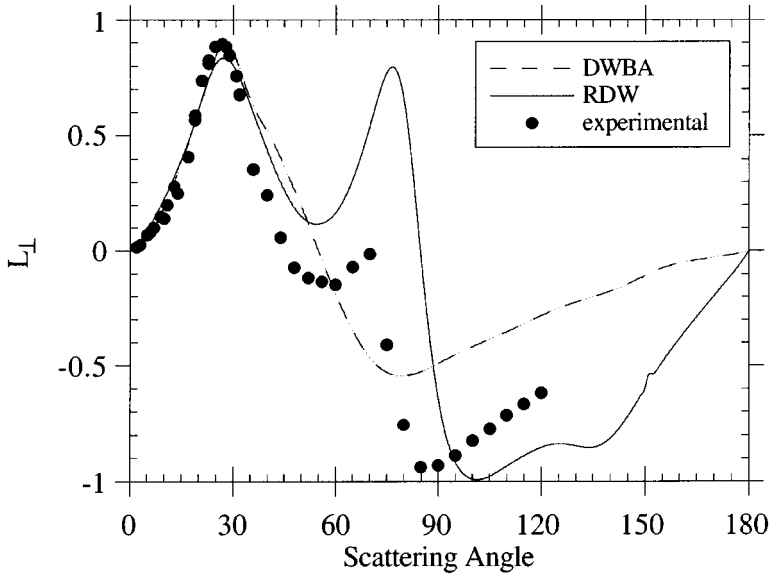

Fig. 2. Measured values of $L_\perp$ for excitation of the $5^2S_{1/2}$–$5^2P_{3/2}$ transition of rubidium by electrons of incident energy 20 eV. The data are an average of that taken for both isotopes and are compared with two distorted wave Born approximation theoretical models, one incorporating a relativistic wave function (RDW, Zeman *et al.* 1998) and one which does not (DWBA, Pangantiwar and Srivastava 1988). The angular resolution of the apparatus has been convoluted with the two theories for comparison with the experimental data.

eV for the de-exciting superelastic scattering experiment. Each point represents a weighted average of data measured from the two isotopes which should yield the same collision parameter value since the nuclear spin is expected to play no role in the collision process (Percival and Seaton 1958). The statistical standard deviation is smaller than the diameter of a plotted point. The only difference found between the two isotopes manifests itself in the optical pumping within their respective hyperfine structure. This difference appears in the respective values of the optical pumping parameters K'. By recording measurements from each isotope, we were able to confirm the Percival–Seaton assumption and the accuracy of the QED modelling of the laser excitation to within experimental error.

The measured data are compared with calculations from two theoretical models using a first order distorted wave Born approximation. One of the calculations (DWBA) includes non-relativistic wave functions (Pangantiwar and Srivastava 1988), while the other (RDW) incorporates a relativistic wave function (Zeman *et al.* 1998). The effects of the finite angular resolution of the apparatus have been taken into account in the theoretical plots by convoluting an appropriate Gaussian function which matches this resolution with the theoretical data.

At scattering angles greater than 30°, neither theoretical calculation agrees well with the experimental data. However, the shape of the RDW model is similar to the measurements, while the DWBA calculations yield a simpler curve

that is like those for the light atoms H and He. The experimental data display the multiple sign changes seen in the potassium data of Stockman *et al.* (1998), although the intermediate minimum and maximum are not as pronounced. This could be due to the difference in incident electron energy used in each experiment and it would be interesting to repeat the rubidium experiment for the potassium conditions of 10 eV incident electron energy.

One reason for studying the heavier rubidium atom was to determine whether relativistic effects are important in the interaction between the projectile electron and the atomic core. Neither theoretical model, whose calculations are shown in Fig. 2, includes any relativistic contribution in the interaction potential. Of significance, however, is that the model incorporating the relativistic wave

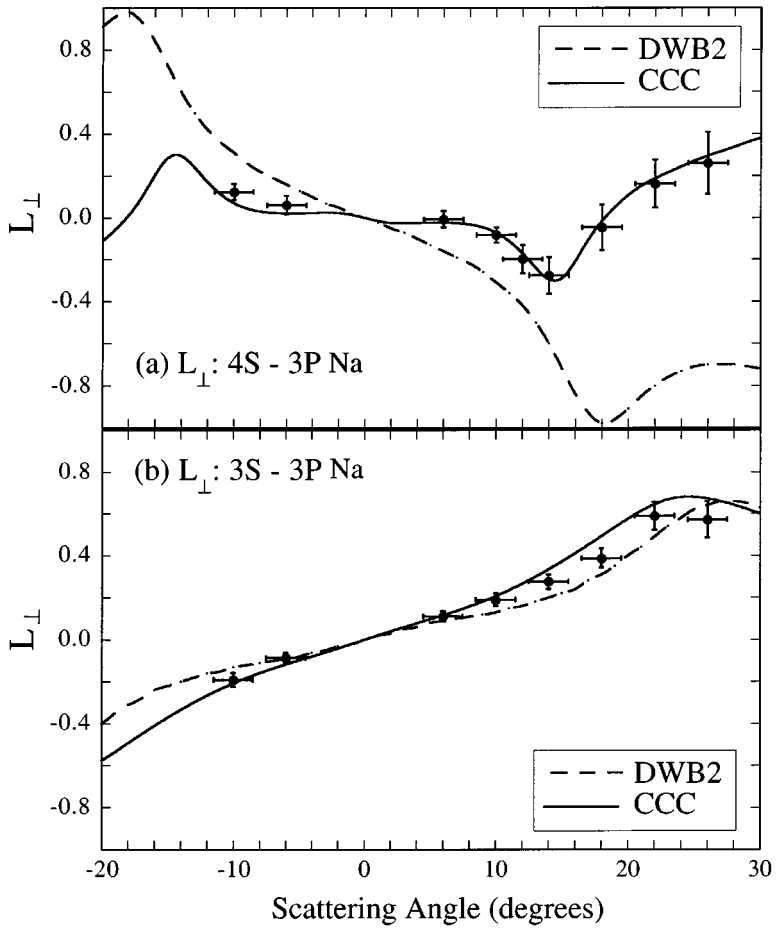

Fig. 3. Measurements of $L_\perp$ for collisions between electrons of incident energy 22 eV and the laser-prepared $3^2P_{3/2}$ level of sodium: (a) de-excitation of the $4^2S_{1/2}$ level and (b) excitation of the $3^2S_{1/2}$ level. In each case, the data are compared with calculations from convergent close coupling (CCC, Bray 1994) and the second order distorted wave Born approximation (DWB2, Madison *et al.* 1991; Bubelev *et al.* 1996) models.

functions displays some qualitative agreement with the experimental data. There is now a need for a theoretical model that addresses the relativistic interaction. A model that has had success in reproducing measured data for electron excitation of light, non relativistic atomic transitions at medium to low electron energies is the non-perturbative convergent close coupling (CCC) theory (Bray and Stelbovics 1992). It is expected that a version of this model able to handle the relativistic interactions will be available soon, allowing comparison with these experimental data.

Fig. 3*a* shows measurements of $L_\perp$ for the de-excitation of the $4^2S_{1/2}$ level of sodium to the $3^2P_{3/2}$ level by electrons of 22 eV incident energy. The results of the superelastic measurement of the $3^2S_{1/2}$–$3^2P_{3/2}$ transition excited by electrons of the same energy and recorded in the same experiment are given in Fig. 3*b*. The measurement of this parameter was part of a more comprehensive study which yielded all the spin averaged ACPs for the 4S–3P transition (Shurgalin *et al.* 1998). Results of the calculations of a second order distorted wave Born approximation (DWB2) (Madison *et al.* 1991; Bubelev *et al.* 1996) and of the CCC model are shown in both cases for comparison. While both theories provide acceptable agreement with the measured data for the lower transition, the CCC displays clearly superior agreement for the 4S–3P transition.

It is also apparent that, for small values of θ, there is a sign reversal for $L_\perp$ when the energy transfer direction between the electron and the atom is reversed. This is in accordance with the hypothesis of Andersen and Hertel (1986) that $L_\perp$ should be negative. However, after decreasing to approximately $-0\cdot25$ at a scattering angle of 15°, $L_\perp$ increases in value becoming positive at about 19°. This behaviour is not predicted by the qualitative argument but is well replicated by the CCC model. It is curious that the DWB2 model is able to predict $L_\perp$ for the 3S–3P transition, whereas it is so poor for the 4S–3P transition. Since the energy transfer is smaller for the 4S–3P transition, one might expect the DWB2 to be able to produce good agreement as it is a perturbative model.

In this work, we have extended the experimental horizon for angular momentum data by presenting measurements for rubidium, the heaviest alkali studied yet, and for a transition between excited states. While the CCC model yields excellent agreement in the latter case, more theoretical development is needed to include relativistic interaction potentials before comparison will be possible with the rubidium data.

Acknowledgments

This work has been supported by the Australian Research Council. BVH wishes to acknowledge the receipt of an Australian Postgraduate Award and MS a Griffith University Postgraduate Research Award.

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Manuscript received 23 November 1998, accepted 30 March 1999