Scattering of Polarized Electrons from Optically Pumped Sodium Atoms

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We have measured the spin-dependence for elastic and superelastic scattering of spin-polarized electrons from optically prepared sodium atoms at selected incident energies from 1.0 to 54.4 eV and for scattering angles ranging from 10° to 140°. Results are presented in terms of physical parameters which emphasize the effects of exchange during collision. The data are compared with predictions from theoretical electron scattering calculations.

Collisions between electrons and atoms or molecules are among the most basic of all physical processes, forming the basis for a wide range of physical phenomena, from electron transport in gaseous discharges, to energy transport in tokomak plasmas, to the chemistry of the earth's upper atmosphere. Accurate modeling of all such processes depends critically on the availability of reliable electron scattering cross sections. Despite the significant advances in our theoretical understanding of electron collision phenomena, it is still often the case that even the most advanced theoretical scattering calculations fail in their ability to reproduce the results of experimental measurements.

A rather large number of experimental techniques have been developed which make use of quantum state preparation and detection in order to probe collision processes with considerable depth. For the particular case of electron–atom collisions, the substantial progress in the use of coherence, correlation, coincidence, polarization, optical pumping and step-wise excitation techniques (Kessler 1985, Andersen *et al.* 1988, Kleinpoppen 1988, MacGillivray and Standage 1988, Hanne 1988, Raith 1988, Kelley 1989, Slevin and Chwirot 1990) has made possible investigations of collision phenomena which are increasingly detailed in their characterization of electron collision phenomena. The ultimate goal of these measurements has been the "complete" or "perfect" experiments envisioned by Bederson in 1969, in which all quantum observables are under direct experimental control. Such measurements provide the most detailed and complete characterization possible of the interactions at work in, and the scattering dynamics of, collisions between electrons and atoms.

The experimental program undertaken in our laboratory over the past several years has focused on the scattering of spin-polarized electrons from optically pumped sodium atoms as a model collision system in which to test, with as much detail as possible, collisions between electrons and atoms (Hertel *et al.* 1987, McClelland *et al.* 1987, 1989). Because sodium behaves very nearly like an ideal one-electron atom, our measurements allow us to to study directly the role of exchange in low energy collisions between spin-$\frac{1}{2}$ particles. One significant advantage of our method is the ability to perform these spin-dependent measurements for both elastic and inelastic scattering processes.

In order to introduce the most important concepts, we first discuss exchange in the elastic scattering of a spin-polarized electron from a spin-polarized one-electron atom. Two independent complex scattering amplitudes are required to completely characterize this scattering process (Kessler 1985). Because the composite spin of the collision is conserved, we choose to denote these two amplitudes as S and T, for scattering in the singlet or triplet composite spin channels, respectively. One primary effect of exchange is to introduce differences, often profound, between these two scattering amplitudes.

A complete characterization of the scattering process would require the determination of only three real parameters, *e.g.* the magnitudes of S and T and the phase difference between them. The absolute phases of S and T are not observable quantities. The cross sections for scattering in either the singlet or triplet spin channel are simply proportional to the squared magnitudes of S or T,

$$\sigma_S \propto |S|^2 \quad \text{and} \quad \sigma_T \propto |T|^2 . \tag{1}$$

In all of our measurements, we have control over the relative spin orientation of the incident electrons and atoms and measure independently the scattering rates $I_{\uparrow\uparrow}$ and $I_{\uparrow\downarrow}$, for incident spins parallel or antiparallel. These rates are related to the singlet and triplet cross sections by

$$I_{\uparrow\uparrow} \propto \sigma_T \quad \text{and} \quad I_{\uparrow\downarrow} \propto \frac{1}{2}(\sigma_S + \sigma_T) \tag{2}$$

In this and all following discussion, we specifically assume that both the electrons and atoms are completely polarized. One can fully account for the effects of incomplete polarization in a straightforward manner without affecting in any significant way either our analysis or the conclusions drawn from our results (Kessler 1985, Hertel *et al.* 1987, McClelland *et al.* 1989).

The conventional spin-unpolarized elastic scattering cross section, denoted by σ_{tot}^{elas}, is given by

$$\sigma_{tot}^{elas} = \frac{1}{4}(\sigma_S + 3\sigma_T) \propto \frac{1}{2}(I_{\uparrow\uparrow} + I_{\uparrow\downarrow}) . \tag{3}$$

Because we are primarily interested in the role of exchange during the collisions, we choose to concentrate on how the cross sections σ_S and σ_T differ as a result of exchange rather than on the cross sections themselves. To this end, we define an exchange asymmetry, A_{ex}, as the difference between the two experimentally observed scattering signals, $I_{\uparrow\uparrow}$ and $I_{\uparrow\downarrow}$, normalized to their sum. That is,

$$A_{ex} = \frac{I_{\uparrow\downarrow} - I_{\uparrow\uparrow}}{I_{\uparrow\downarrow} + I_{\uparrow\uparrow}} = \frac{\sigma_S - \sigma_T}{\sigma_S + 3\sigma_T} . \tag{4}$$

This exchange asymmetry is directly related to to the ratio r between the triplet and singlet cross sections by

$$r = \frac{\sigma_T}{\sigma_S} = \frac{1 - A_{ex}}{1 + 3A_{ex}} . \tag{5}$$

One important advantage of measuring relative quantities, such as the exchange asymmetry or the cross section ratio, is that one can extract directly from the experimental observations specific information about a specific effect, in this case exchange, without suffering from the numerous systematic problems which plague accurate measurements of absolute scattering cross sections.

In the limit of a vanishing triplet scattering cross section, A_{ex} takes on a value of +1 and the cross section ratio is zero. If, however, it is the singlet cross section that becomes very small, then A_{ex} takes a value of $-\frac{1}{3}$ and the ratio approaches infinity. If exchange plays a negligibly small role in the collision, then S and T are identically equal so that A_{ex} vanishes and the ratio is unity.

Because the triplet/singlet cross section ratio, or equivalently the exchange spin asymmetry, measures how σ_S and σ_T differ from the spin unpolarized cross section σ_{tot}^{elas}, a measurement of either A_{ex} or r, along with an absolute measurement of σ_{tot}^{elas}, is sufficient for a determination of both $|S|$ and $|T|$.

We now turn to a discussion of exchange in inelastic collisions. Such collisions can be described by a straightforward generalization of the ideas used to describe elastic scattering. In our experiments, we are interested in collisions which involve transitions between the $3S$ and $3P$ states in sodium (Hertel *et al.* 1987, McClelland *et al.* 1989). In addition to the independent spin channels corresponding the two composite spin states of the coupled spin-$\frac{1}{2}$ system, there are two independent orbital angular momentum channels,† corresponding to excitation of the $M_L = +1$ and -1 magnetic sublevels. Thus, a total of four complex scattering amplitudes are required to describe this process completely. We label these S_{+1}, S_{-1}, T_{+1} and T_{-1}, for excitation of the +1 and −1 magnetic sublevels through either the singlet or triplet composite spin channels.

In order to determine the magnitudes of these four scattering cross sections, one needs to measure three quantities in addition to the conventional unpolarized inelastic differential scattering cross section. We choose these to be the ratio r between the triplet and singlet scattering cross section, averaged over excitation of the $M_L = \pm 1$ magnetic sublevels, and two parameters, referred to as the angular momentum transfer parameters, which characterize the differences between the excitation of the $M_L = +1$ and $M_L = -1$ magnetic sublevels via scattering in either the singlet or triplet spin channel. These parameters, along with the conventional scattering cross section, are related to the four complex scattering amplitudes by

$$\sigma_{tot}^{inelas} = \frac{1}{4}\left(|S_{+1}|^2 + |S_{-1}|^2 + 3\,|T_{+1}|^2 + 3\,|T_{-1}|^2\right) \tag{6}$$

$$r = \frac{|T_{+1}|^2 + |T_{-1}|^2}{|S_{+1}|^2 + |S_{-1}|^2} \tag{7}$$

$$L_\perp^S = \frac{|S_{+1}|^2 - |S_{-1}|^2}{|S_{+1}|^2 + |S_{-1}|^2} \tag{8}$$

$$L_\perp^T = \frac{|T_{+1}|^2 - |T_{-1}|^2}{|T_{+1}|^2 + |T_{-1}|^2}. \tag{9}$$

† Because the collision conserves the overall positive reflection symmetry about the scattering plane, any angular momentum transferred during the collision must be normal to that plane. As a consequence, in the coordinate system in which angular momentum is quantized along the scattering plane normal, the amplitude for excitation of the $M_L = 0$ magnetic sublevel must vanish.

As with the elastic scattering measurements, we concentrate on measurements of the relative quantities r, $L_{\perp}^{S}$ and $L_{\perp}^{T}$, which characterize the differences in size between the four complex scattering amplitudes.

One could determine these quantities through observation of an inelastically scattered electron in coincidence with the fluorescence photon subsequently emitted by the atom after excitation by electron impact (Slevin 1990). We choose, rather, to measure the spin dependence through superelastic scattering, whereby one first prepares, by laser optical pumping with circularly polarized light, a population of atoms oriented in the first excited state, and then detects only "superelastic" electrons which have de-excited the atoms, thereby gaining the 2.1 eV excitation energy of the 3P atoms. Because of time reversal symmetry, both inelastic and superelastic scattering are described by the same complex scattering amplitudes.

The relative quantities r, $L_{\perp}^{S}$ and $L_{\perp}^{T}$, are derived in a straightforward way from the four experimental intensities $I_{\uparrow\uparrow}$, $I_{\uparrow\downarrow}$, $I_{\downarrow\uparrow}$ and $I_{\downarrow\downarrow}$, corresponding to the four possible combinations for the polarization directions of the incident electrons and atoms (Hertel *et al.* 1987, McClelland *et al.* 1989).

Our experimental apparatus, a detailed description of which was given in McClelland *et al.* 1989, is shown schematically in Figure 1. Spin-polarized electrons, produced by photoemission from a GaAs photocathode (Pierce *et al.* 1980), are scattered from a collimated beam of sodium atoms. The photoemitted electrons have a net spin polarization of about 32%, oriented either normal or anti-normal to the crystal surface depending on the helicity of the incident circularly polarized laser light. The photoemitted electrons are collected and deflected by 90° to form a beam with transverse spin polarization, either up or down in the laboratory. The energy of the beam is variable from about 1 to 100 eV and is focused to a nominal diameter of 2 mm at the scattering center.

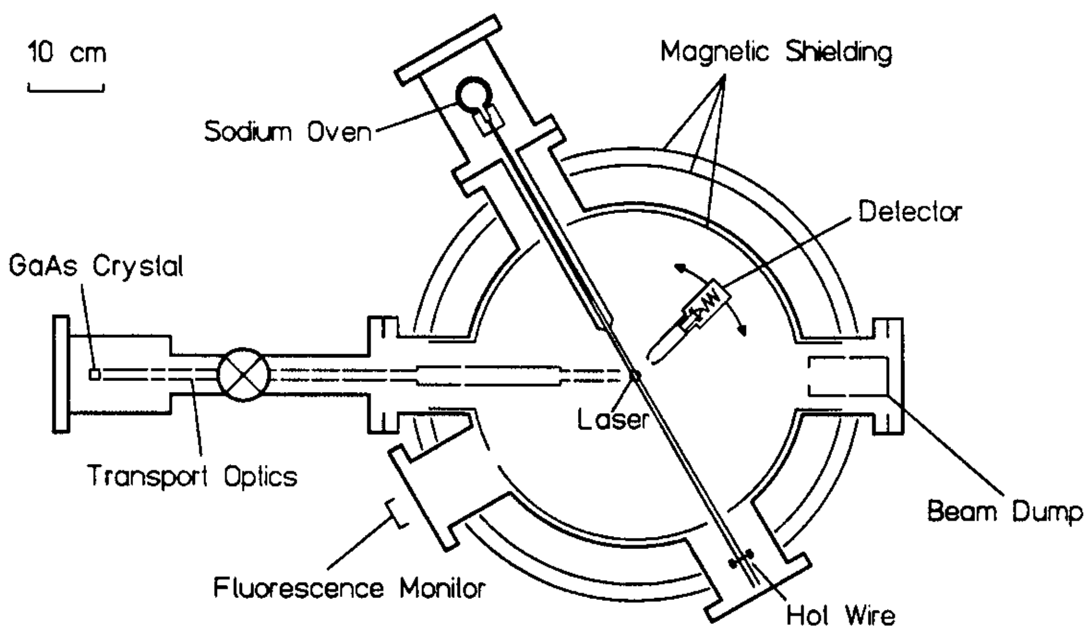

Figure 1: Schematic drawing of the polarized-electron — polarized-atom scattering apparatus.

The beam of sodium atoms is collimated from a continuously recirculating, effusive oven to form a beam of about 4 mm diameter at the scattering center. The target density is about 5×10^9 atoms/cm^3. The atoms are prepared by optically pumping simultaneously at two frequencies corresponding to two hyperfine transitions in sodium. The atomic polarization direction, again either up or down in the laboratory, is determined by the helicity of the circularly polarized laser light used for optical pumping. For elastic scattering measurements, the optical pumping is performed just slightly upstream in the atom beam from the scattering center, so that only ground state atoms are present in the target region. The net spin polarization of the ground state atoms is about 98%. For the superelastic measurements, the optical pumping takes place in the scattering center and approximately 50% of the atoms can be maintained in the $3P$ excited state by the laser excitation. In addition to having essentially 100% spin polarization, the excited atoms are maintained in pure magnetic sublevels with $M_L = +1$ or -1, depending on the helicity of the pumping light.

Electrons scattered through an angle θ_{scat}, in the range from about 15° to 140°, are collected at the detector. The detector includes a retarding field energy analyzer for discrimination against inelastic scattered electrons in the case of elastic scattering measurements, and against both elastic and inelastic scattered electrons in the case of superelastic scattering measurements. For both the elastic and superelastic measurements, the scattered signals are accumulated for each of the four possible relative spin orientations of the incident electrons and atoms. From these four scattering signals we construct the parameters by which the role of exchange is characterized (Hertel *et al.* 1987).

Results of our measurements of spin dependence in elastic and superelastic electron scattering from sodium are shown in Figures 2 and 3 (McClelland *et al.* 1985, 1986, 1989, Buckman *et al.* 1989). The uncertainties shown in all figures are derived by propagation of the one standard deviation uncertainty due to counting statistics through the expression for the observed quantity. Uncertainties are shown only where they exceed the size of the plotted symbols.

In Figure 2 we show our measurements of the elastic exchange asymmetry at incident energies from well below the first excitation threshold up to about ten times the ionization potential for sodium. The measurements shown in Figure 2a and 2b, at 1.0 and 1.6 eV incident energy, respectively, are the first such detailed measurements of elastic scattering below the first excitation threshold. In both cases, the predominantly negative exchange asymmetry indicates that, over most of the angular range, triplet dominates over singlet scattering by as much as a factor of two. Only at scattering angles larger than 90° does singlet scattering become comparable to triplet scattering.

At these energies the elastic channel is the only open scattering channel, so one would expect that a close-coupling approximation should provide a reasonably accurate calculation for the elastic scattering. Indeed, we find excellent agreement of the four-state close-coupling calculation from Moores and Norcross (1972) with these experimental results.

As the incident energy increases and more scattering channels open up, corresponding to the possibility for collisional excitation of higher lying atomic levels, one might expect that inaccuracies arising from the neglect of the higher levels in the calculations would degrade the agreement between theory and experiment. Of particular interest, then, is to study how the agreement degrades with increasing incident energy in order to assess the range of validity of calculations which include a limited number of excited states. Figures 2c through 2g show the results of our measurement at increasing in-

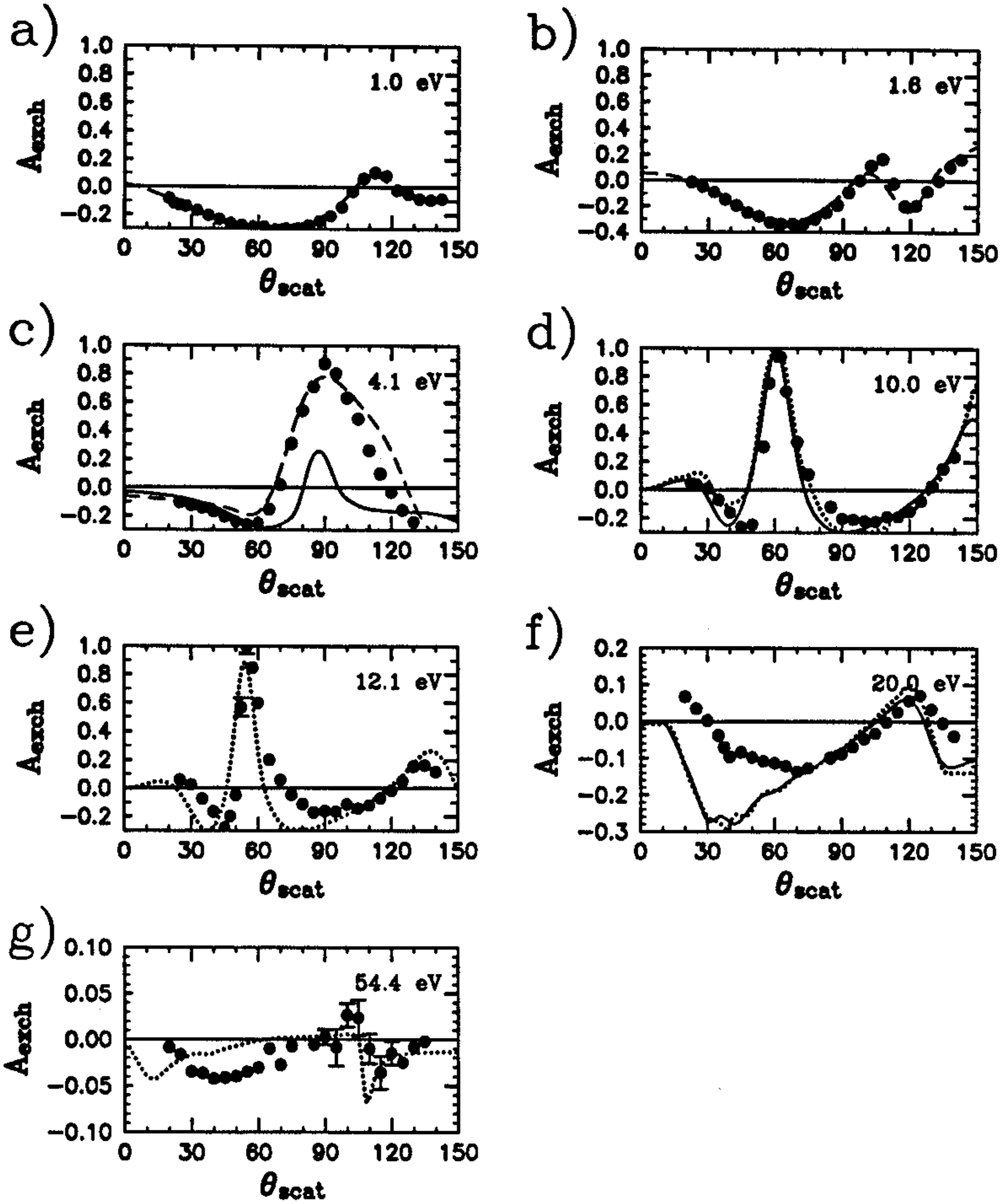

Figure 2: Exchange asymmetry A_{ex} for elastic scattering from sodium. Data points are our experimental results with error bars, one standard deviation derived by propagation of the uncertainty due to counting statistics, shown only when larger than the plotted symbols. Smooth curves represent the results of four-state close-coupling calculations: (– – –) Moores and Norcross (1972) (Figures a, b and c); (——) Oza (1988) (Figures c, d and f); (· · · ·) Mitroy *et al.* (1987) (Figures d, e, f and g).

cident energies. As shown in Figure 2c for scattering at 4.1 eV, the character of the exchange asymmetry has changed significantly, with singlet scattering dominating over triplet scattering by about a factor of five in the region near 90°. At 10.0 and 12.1 eV, the large positive peak in the spin asymmetry has moved toward smaller scattering angles and has narrowed significantly. At 20.0 eV, this peak is no longer visible and the reduced size of the spin asymmetry overall is indicative of the reduced importance of exchange at this energy. From Figure 2g one notes that by the time the incident energy reaches about 50 eV, exchange plays a minimal role in the collision.

Comparison with theory at the higher energies shows a couple of interesting features. The calculation of Moores and Norcross (1972), shown in Figure 2c as the dashed line, still provides excellent agreement at nearly twice the excitation threshold for sodium. A four-state calculation of Oza (1988), shown as the solid curve in Figures 2c, 2d and 2f, shows rather poor agreement at 4.1 eV, significantly underestimating the relative importance of singlet scattering in the region around 90°. At 10.0 eV, however, Oza's calculation shows excellent agreement with experiment. A four-state calculation of Mitroy *et al.* (1987), shown as the dotted curves in Figures 2d through 2g, is also available and agrees very closely with that of Oza (1987). Both calculations show excellent agreement at 10.0 and 12.1 eV, but are unsatisfactory at 20.0 eV and above.

In addition to elastic exchange asymmetries, the close-coupling calculations provide a detailed characterization of electron impact excitation of the 3P from the ground 3S level in sodium. Thus, it is interesting to assess their reliability for inelastic scattering as well. We present in Figure 2 results of our superelastic scattering measurements for spin dependence in inelastic electron–sodium scattering at total energies from 4.1 to 40.0 eV.[‡]

Several interesting features are readily apparent from these data. First, there are rather striking differences between the singlet and triplet angular momentum transfer parameters $L_\perp^S$ and $L_\perp^T$, particularly at the lower energies. This indicates that exchange has a rather strong influence on the scattering at these energies. It is also apparent that the spin-averaged angular momentum transfer $L_\perp$ bears a much closer resemblance to the triplet angular momentum transferred $L_\perp^T$ than to $L_\perp^S$. This is in large part due to the fact that, in unpolarized measurements, the triplet composite spin state is three times more likely to occur than the singlet state, allowing it to dominate the observed angular momentum transfer.

One also notices that the triplet/singlet ratio deviates quite strongly from unity, another indication of the importance of exchange for collisions at these energies. At all measured energies except 4.1 eV, the triplet scattering cross section appears generally to dominate the singlet channel, often by a considerable amount. At 4.1 eV, singlet scattering dominates by about a factor of two both at large and small scattering angles. For scattering at intermediate angles from about 60° to 90°, scattering in the triplet channel is about 1.5 times stronger than in the singlet.

The development, with increasing scattering energy, of both the angular momentum transfer and the cross section ratio is also interesting. At 4.1 eV, both $L_\perp^S$ and $L_\perp^T$ show a positive maximum at about 40°, with a second positive maximum at larger angles. This second maximum occurs at a larger angle for $L_\perp^S$ than for $L_\perp^T$, and $L_\perp^S$ also has an

[‡]The total energy is the incident energy plus the 2.1 eV excitation energy of the sodium 3P level. We use this energy in order to make connection with theoretical calculations where the scattering energy is given relative to the ground state.

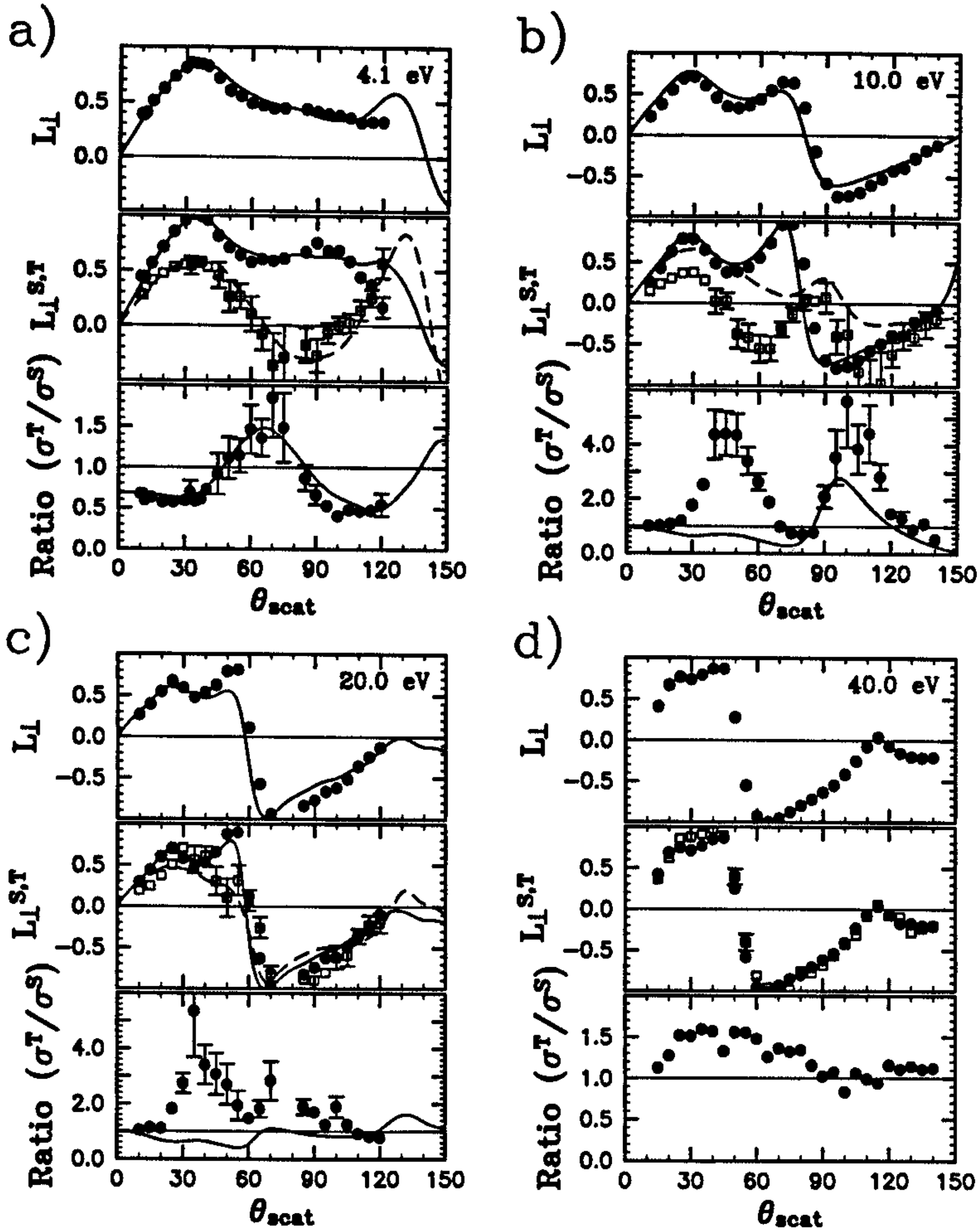

Figure 3: Spin-averaged and spin-resolved angular momentum transfer and the triplet/singlet cross section ratio for collisional excitation of the $3S \rightarrow 3P$ transition. Our measurements are shown as data points. Error bars, one standard deviation due to counting statistics, are shown only when larger than the plotted symbols. $L_\perp^S$ is represented by open squares and dashed lines, while $L_\perp^T$ is represented by the closed circles and solid lines. The theoretical curves are the results of four-state close-coupling calculations. At 4.1 eV the theoretical results are taken from Zhou *et al.* (1991). At 10.0 and 20.0 eV they are taken from Mitroy *et al.* (1987).

intermediate region of negative angular momentum transfer which is not seen for $L_{\perp}^{T}$.

These features are essentially repeated for scattering at 10.0 eV, except that they are compressed towards smaller angles. With the compressed angular dependence, the cross section ratio shows a second prominent peak, around 100°, which was not visible at 4.1 eV. Further, the secondary peak at larger scattering for the angular momentum transfer is more pronounced, particularly for the triplet channel.

While the same features remain visible for scattering at 20.0 eV, though further compressed in angle, the two peaks in the angular momentum transfer have begun to merge and the large angle maximum in the cross section ratio has reduced significantly. At 40.0 eV, the data are still further compressed in angle and the peaks in the angular momentum transfer have nearly completely merged. The singlet and triplet angular momentum transfers are essentially indistinguishable and the triplet/singlet ratio is much reduced, indicating that at this high scattering energy, exchange has little effect on the scattering.

The results of theoretical close-coupling calculations are also shown in Figure 3. Figure 3a shows the results of a seven-state calculation by Zhou *et al.* (1991) which is in excellent agreement with the experimental results. As was also the case with elastic scattering, however, the reliability of the theoretical calculations is worse at the higher energies. The results of Mitroy *et al.* (1987) are shown for scattering at 10.0 and 20.0 eV in Figures 3b and 3c. In both figures, the calculation for the triplet and the spin-averaged angular momentum transfers are quite satisfactory. The calculation for the singlet angular momentum transfer is not quite as accurate, particularly for the 10.0 eV results. The source of the severe breakdown of this calculation for the triplet/singlet cross section ratio at both 10.0 and 20.0 eV is not at present understood. No theoretical results are available for comparison with our measurements at 40.0 eV.

The data presented in this work represent a rather detailed characterization of electron scattering from sodium, both in the elastic and in the dominant inelastic scattering channel. The measurements span a wide range of energies from below the first excitation threshold up to about 10 times the ionization potential, providing a comprehensive body of data which can serve as a benchmark for evaluating the results of sophisticated electron–atom scattering calculations. The close-coupling calculations provide an excellent description of the scattering at very low energies and give promising results at higher energies. It is hoped that the results of this experimental program will stimulate further theoretical work and lead to a deeper understanding of this fundamental physical process.

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