SPIN-POLARIZATION, ORIENTATION, AND ALIGNMENT IN ELECTRON-ATOM COLLISIONS

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The use of state-selection techniques has added greatly to our fundamental understanding of electron–atom collision phenomena. In particular, measurements of atomic orientation and alignment in electron impact excitation, and measurements of spin dependence in both elastic and inelastic scattering, provide substantially more detail about collision processes than conventional measurements of differential cross sections alone. This work reviews recent experiments which, through the use of spin dependence and atomic orientation and alignment, attempt to provide the most complete characterization possible of electron–atom collision phenomena.

1 Introduction

Collision studies which make use of quantum state preparation and detection techniques have led to a considerable advance in our knowledge of scattering phenomena. In particular, for the case of electron-atom collisions, the substantial progress seen in coherence, correlation, coincidence, polarization, optical pumping, and step-wise excitation techniques has made possible experimental investigations which are increasingly detailed in their characterization of scattering processes. In some favorable cases, the truly "complete" or "perfect" measurements envisioned by Bederson[1] are possible. Such measurements provide the most detailed and complete characterization possible of the interactions at work in and scattering dynamics of collisions between electrons and atoms.

Several excellent reviews document the status of experiments utilizing state-selection techniques, including electron–photon coincidence measurements,[2–5] collisions using laser-excited atoms,[6,7] and spin-polarization techniques in electron–atom scattering.[8–11] The breadth of these combined fields of study far exceeds the scope of the present work. We will rather concentrate on several recent experiments which combine elements of each of these fields in order to approach more closely the ultimate goal of "complete" scattering experiments.

We begin in Sec. 2 with the introduction of some general concepts, in the context of inelastic scattering, which are required for the understanding of the experiments to be presented. We follow in Sec. 3 with a discussion of the origin of spin dependence in electron scattering, and describe spin-dependent measurements of elastic electron scattering. Finally, in Sec. 4 we describe several recent experiments which study both the spin dependence and orbital angular momentum alignment in inelastic electron–atom scattering.

2 General Principles of State Selection: S → P Excitation

Let us consider a prototypical electron scattering event in which an incident electron collides with a target atom and scatters to a detector placed at some scattering angle. Our goal is to determine from this measurement as much as possible about this collision process and the interactions at work.

How much could we learn about this collision event? Everything that can be learned about any such collision is contained in the wavefunction of the colliding system, so that's what we should try to determine. One generally describes this wavefunction in terms of complex scattering amplitudes which connect the initial state with each possible final state of the system. Determination of all scattering amplitudes is then equivalent to a determination of the full wavefunction, and provides a complete description of the collision process.

Of course, characterization of the final wavefunction requires knowledge of both the final state of the target atom and the final state of the scattered electron, so we must determine the complete set of scattering amplitudes connecting all initial to all final states. That is, we must study the scattering to all angles, for all combinations of initial and final states of the electron and target atom.

This is a rather ambitious undertaking, but is exactly what is necessary for a "complete" experiment. For essentially all experiments to date, however, one has been content to determine the set of scattering amplitudes for a particular scattering channel, e.g. for elastic scattering or for some particular atomic excitation. While such measurements are in a limited sense "complete," we should not lose sight of the ultimate objective of determining the scattering amplitudes for all processes at work during the collision.

Even in the more restricted sense of "complete" experiments for a single resolvable scattering channel, there are two immediate conceptual problems with the wavefunction determination as proposed above. First, it is not possible to determine directly the phase of the complex valued wavefunction. It would thus seem that at least the phases of the scattering amplitudes would be inaccessible. Second, it is generally not possible to ensure that either the initial or final state of the system is a single, well-defined quantum state, as there are typically degenerate magnetic sublevels which cannot be resolved by conventional techniques. The goal of much of the effort invested in coherence, correlation, coincidence, polarization, and step-wise excitation techniques has been to develop methods with which to overcome these difficulties.

Let us take the illustrative example of electron impact excitation from an S to a P state, ignoring any effects of electron spin. The S state has only the single $M_L = 0$ magnetic sublevel, but the P state has three degenerate sublevels, $M_L = \pm 1, 0$. Thus, three scattering amplitudes, f_{+1}, f_{-1}, and f_0, are required to describe this excitation. However, because of the overall positive reflection symmetry about the scattering plane (as defined by the incident and scattered electron momenta), these three are not all independent and only two need to be determined.

How these three amplitudes reduce to two is dependent upon the choice of coordinate system. Because one result of the reflection symmetry is that angular momentum transferred in the collision must be perpendicular to the scattering plane, it is convenient to choose the normal to the scattering plane as the quantization axis for the atomic angular momentum. In this coordinate system, the so-called "natural" frame, excitation of the $M_L = 0$ level is forbidden, so the f_0 amplitude vanishes exactly. Only the amplitudes f_{+1} and f_{-1} need to be determined.†

It is in principle straightforward to determine the magnitudes of f_{+1} and f_{-1}. This can be done in alkali atoms, for example, by collisional de-excitation of atoms maintained in the first excited state by laser optical pumping. One knows from time reversal symmetry that the same amplitudes must describe both the excitation and the de-excitation processes. By pumping with circularly polarized light incident perpendicular to the scattering plane, one can prepare excited atoms in fully oriented, pure magnetic sublevels of the P state, either $M_l = +1$ or $M_L = -1$. From the scattering cross section for collisional de-excitation of these P atoms, which is simply proportional to $|f_{+1}|^2$ or $|f_{-1}|^2$, one determines the magnitude of the amplitudes, $|f_{+1}|$ and $|f_{-1}|$.

Rather than determine $|f_{+1}|$ and $|f_{-1}|$ separately, it is generally preferable to determine the conventional differential cross section, proportional to $|f_{+1}|^2 + |f_{-1}^2|$, and a relative measure of the difference between $|f_{+1}|$ and $|f_{-1}|$. One example of such relative measurements is $L_\perp$, the net orbital angular momentum which would be transferred to the P state via electron impact excitation from the ground state, defined by

$$L_\perp = \frac{I_{+1} - I_{-1}}{I_{+1} + I_{-1}} \tag{1}$$

where $I_{\pm 1}$ refer to the scattering signals for exciting or de-exciting the $M_L = \pm 1$ sublevels. Because the experimental signals appear both in the numerator and denominator of this expression, experimental factors such as target densities, detector efficiencies, and beam fluxes are completely canceled out. From a determination of the conventional differential cross section and $L_\perp$, or R, one can determine both $|f_{+1}|$ and $|f_{-1}|$.

It is in principle also straightforward in this case to determine the phase difference between these two scattering amplitudes from a study of the collisional de-excitation of atoms pumped with linearly polarized light, again incident normal to the scattering plane. Atoms excited with linearly polarized light have a "peanut"-shaped charge distribution, and the cross section for de-excitation depends on the alignment of this "peanut" with respect to the direction of incidence of the incoming electron. A determination of the alignment angle which maximizes (or minimizes) the cross section gives a direct measure of the phase difference between the two scattering amplitudes.

The connection between this alignment angle and the relative phase of the scattering amplitude is readily apparent if one thinks of the "peanut"-shaped P state as a linear superposition of the $M_L = \pm 1$ sublevels. That is, one thinks of the "peanut" as a standing wave generated by the interference between the two counter-rotating "doughnut"-shaped charge clouds of the $M_L = \pm 1$ sublevels. The relative phase of the two "doughnuts" determines the alignment angle of the "peanut".

The scattering from each of the $M_L = \pm 1$ sublevels contributes coherently to the observed scattering signal, generating an "interference pattern" at the detector which, in this picture, gives rise to the sinusoidal dependence of the scattering cross section on the alignment angle. The phase of this "interference pattern", i.e. the angle at which the maximum scattering occurs is determined by both the relative phase between the two initial states and the phase difference between the

†Another common coordinate system is one in which the incident direction of the projectile electron serves as the quantization axis, the so-called "collision" frame. In this system, the $M_L = \pm 1$ sublevels cannot independently be excited and the corresponding scattering amplitudes have a fixed relationship with each other, i.e. $\mathcal{F}_{+1} = -\mathcal{F}_{-1}^*$. One thus needs to determine the two amplitudes $\mathcal{F}_0$ and $\mathcal{F}_1$.

two scattering amplitudes. Consequently, determination of phase of the interference pattern gives one a direct measurement of the phase difference between the two scattering amplitudes.

It is important to point out, however, that only through the lack of degeneracy in the final S state was a complete determination of the amplitudes possible with no explicit analysis of the atomic state after scattering. In the more general case, where multiple initial and final states exist, state selection is required both before and after the collision in order to make a full determination of scattering amplitudes.

This principle of separately determining the amplitudes and phases of the scattering amplitudes, as described above, applies equally well to any other scattering process and is readily generalized to collision systems involving state degeneracy for both the initial and final states of the electron and target atom. Independent of what the scattering process might be, one chooses a suitable coordinate system and basis set with which to describe the collision system. Then, one only needs to measure the scattering cross section for each pair of incident and final states, and to determine, in a pair-wise fashion, the interferences between scattering channels in order to completely determine the magnitudes and phases of all scattering amplitudes, apart from one overall phase.

Of course, this idealized measurement protocol can rarely be achieved in practice. Nevertheless, the conceptual description is very useful and is the fundamental idea underlying the density matrix formalisms commonly used to handle the complicated bookkeeping involved in describing complicated collision processes involving techniques of state preparation and detection. There has been much effort directed towards developing general formulae for relating calculated scattering cross sections to quantities observed in state-selected measurements.[12,13] We will not be concerned here with specific formulae but will refer the reader to the original publications of individual experiments for further details.

For the present we merely note that for essentially all experiments, results are reported in terms of physical parameters, analogous to the angular momentum transfer, $L_\perp$, discussed above, which characterize some physical aspect of the collision or measurement process and are related in a known way to the underlying scattering amplitudes. For the case of electron–photon coincidence studies of atomic excitation, results are generally reported in terms of the shape of the charge cloud for the excited atom ($L_\perp$, P_{lin}, γ, and ρ_{00},[4] or alternatively λ, $\overline{\chi}$, Δ, and ϵ,[14,15]) or the state multipoles of the density matrix which describes the excited state.[13]

3 Spin-Dependence in Elastic Electron Scattering

The preceding description of inelastic $S \rightarrow P$ transitions would be complete if electrons had no spin. Of course they do, and the study of spin dependence in electron scattering has proven valuable for furthering our understanding of fundamental aspects of electron–atom scattering. Besides the additional information about scattering dynamics available from studies of the spin dependence, one can use spin dependence to aid in classification of unknown resonance features in elastic or inelastic scattering and to study directly optically-forbidden, spin-changing transitions which are otherwise difficult to observe.[8,10,11]

In order to interpret the results of any experiment involving polarized electrons, it is useful first to consider separately the two underlying causes of spin dependence in elastic electron scattering — the spin-orbit interaction and exchange. For each, as for the case of inelastic $S \rightarrow P$ transitions, one chooses a basis set which simplifies the understanding of the corresponding pure-state to pure-state collision events. A determination of the corresponding cross sections and pairwise interferences then comprises a complete measurement.

3.1 The Spin-Orbit Interaction

The spin-orbit interaction arises from the interaction of the magnetic moment (spin) of an incident electron with the magnetic field observed in the rest frame of that electron due to its motion in the electric field of the scattering target. This effective magnetic field is always normal to the scattering plane determined by the incident and scattered electron momenta, so that electrons at a given impact parameter whose spins are "up", relative to the scattering plane, have an energy which is different from that of electrons whose spin is "down". The scattering of spin "up" and spin "down" electrons is thus determined by different scattering amplitudes, say $f_\uparrow$ and $f_\downarrow$, and hence different scattering cross sections.

This difference between the scattering cross sections leads to both of the primary observables for spin-orbit scattering. First, it causes the left/right scattering asymmetry, S_A, which is exploited in Mott detectors for the determination of electron spin. Second, it causes electrons with no initial spin component normal to the scattering plane to acquire a normal component, S_P, after scattering from heavy atoms. For most cases studied in elastic scattering, where the target atom has no net angular momentum, $S_A = S_P$ so that observation of either asymmetry, along with the spin-averaged differential cross section, is sufficient to determine $|f_\uparrow|$ and $|f_\downarrow|$.

The phase difference between $f_\uparrow$ and $f_\downarrow$ can be determined in a measurement analogous to that use for the $P \rightarrow S$ de-excitation described in Sec. 2. One prepares an initial ground state which is a superposition of "up" and "down", i.e. polarized in the scattering plane, and measures the phase of the resulting "interference pattern", i.e. the net precession angle of the spin about the scattering plane normal. This net precession angle is directly related to the phase difference between $f_\uparrow$ and $f_\downarrow$. Thus, complete characterization of elastic spin-orbit scattering is achieved by determining the spin polarization after scattering of an electron initially polarized in the scattering plane.

Rather than using the amplitudes $f_\uparrow$ and $f_\downarrow$ which we have just described for the scattering of "up" and "down" electrons, one conventionally uses the amplitudes f and g for "direct" and "spin-flip" scattering.[8] These pairs of amplitudes are related by

$$f_\uparrow = f - ig \qquad f_\downarrow = f + ig. \tag{2}$$

There have been many studies of spin asymmetries in elastic scattering which have added significantly to our understanding of scattering from heavy atoms.[8–10] We take as an example work from the Münster group for electron scattering from xenon and mercury in which a complete set of such measurements were made.[16–19]

The essential features of the apparatus used by the Münster group are a source of electrons which are spin-polarized in the scattering plane, an atomic beam scattering target, and an electron spin polarimeter. The spin polarimeter determines only two transverse spin components of the scattered electron, but a Wien filter was optionally used to rotate the longitudinal spin component of the scattered electron into the transverse direction so that a complete determination of all three components could be made.

The measurement of the spin polarization after scattering, together with a measurement of the absolute spin-averaged differential scattering cross section comprises a determination of the complete set of observables for this scattering process. Consequently, Berger, *et al.* were able to determine directly both the magnitude and relative phase of the two complex scattering amplitudes f and g. We show in Fig. 1 their results for scattering from Xe at 50.0 eV incident energy.

The striking difference between the angular dependences for $|f|$ and $|g|$ stems from the rather fundamental difference between the contributions of f and g to the scattering cross section. The conventional spin-averaged differential cross section is largely determined by f, with g, typically an order of magnitude smaller than f, adding a rather small correction due to the spin-orbit interaction. This interaction is strongest in the high electric field near the nucleus and is thus most important at small impact parameters. Consequently, only the lowest few partial waves should contribute to g, as is reflected by its rather smooth angular dependence.

Also shown in Fig. 1 are results of theoretical calculations from various groups.[20–22] The overall agreement is quite good at this as well as the other incident energies studied. Similar agreement was also found for electron scattering from mercury, testifying to the ability of current scattering calculations to model this scattering process accurately.

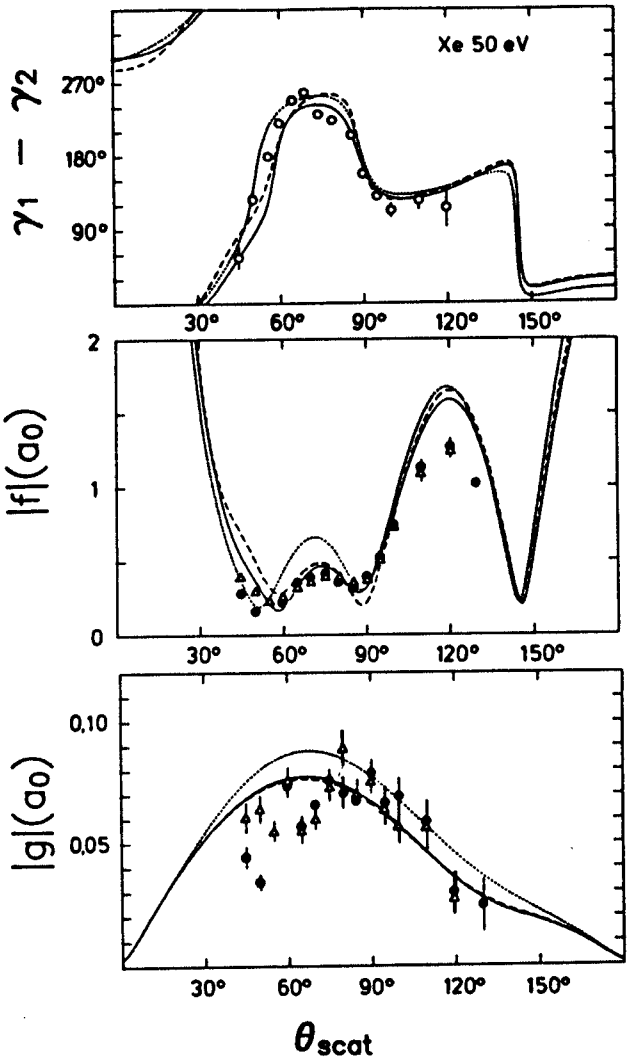

Fig. 1. Scattering amplitudes for Xe at 50 eV incident energy. Experimental data from Berger, *et al.*[18] For the determination of $|f|$ and $|g|$, the absolute cross section measurements of Register, *et al.*[23](•) and Mehr[24](Δ) were used. Theoretical curves are from Haberland, *et al.*[20](——), McEachran and Stauffer[21](· · · ·), and Awe[22](- - - -).

3.2 Exchange Scattering

The other source of spin dependence, exchange, differs from the spin-orbit interaction, in that it is not the result of any spin-dependent force at work during the scattering, but is a manifestation of inherent symmetry properties of the wavefunction for spin-$\frac{1}{2}$ particles. The simplest case to consider is the scattering of a polarized electron from a polarized one-electron atom such as hydrogen, or equivalently, an alkali. These systems provide a particularly good example for the study of exchange because the dependence of the scattering cross section on the relative spin orientation of the incident electron and atom gives a direct measure of the role of exchange in the collision.

Neglecting the spin-orbit interaction, there are only three possible non-equivalent collision events:

$$e(\uparrow) + H(\downarrow) \rightarrow e(\uparrow) + H(\downarrow) \quad (3)$$

$$e(\uparrow) + H(\downarrow) \rightarrow e(\downarrow) + H(\uparrow) \quad (4)$$

$$e(\uparrow) + H(\uparrow) \rightarrow e(\uparrow) + H(\uparrow) \quad (5)$$

In the first example, the spins of both particles remain unchanged, so one can say that no "exchange" has occurred, and the scattering is characterized by a "direct" scattering amplitude f.[‡] In the second example, both spins have changed, so "exchange" has clearly taken place, and the scattering is characterized by the "exchange" amplitude $-g$. In the third process, one cannot tell whether the electrons have "exchanged" or not, so both channels contribute and the scattering is described by the amplitude $f - g$.

While this description parallels closely the measurements one can actually make, it does not clearly reflect the inherent symmetry of spin-$\frac{1}{2}$ particles. This symmetry is emphasized by describing the two particle wavefunction in a basis set in which the two spins have been coupled to form singlet and triplet states. Because we have assumed no spin-orbit interaction, the total spin of the colliding system is conserved, and these singlet and triplet states are eigenstates of the scattering Hamiltonian and form independent, non-intermixing scattering channels. Each is described by a complex scattering amplitude, which we label S or T, having the following relationship with the "direct" and "exchange" amplitudes:

$$S = f + g \qquad f = \tfrac{1}{2}(S + T) \quad (6)$$

$$T = f - g \qquad g = \tfrac{1}{2}(S - T) \quad (7)$$

Several possible approaches to the determination of the spin-dependent information are apparent from the scattering scenarios shown in Eqs. 3–5. For example, consider experiments in which both incident particles are spin-polarized, but for which no spin analysis of the scattered particles is performed. In this case, two scattering intensities can be measured which are proportional to the scattering cross sections for the incident parallel and antiparallel relative spin orientations. These cross sections are $\sigma_{\uparrow\uparrow} = |T|^2$ and $\sigma_{\uparrow\downarrow} = \frac{1}{2}(|S|^2 + |T|^2)$, respectively. One can define an exchange asymmetry, A_{ex}, by

$$A_{ex} = \frac{\sigma_{\uparrow\downarrow} - \sigma_{\uparrow\uparrow}}{\sigma_{\uparrow\downarrow} + \sigma_{\uparrow\uparrow}} = \frac{|S|^2 - |T|^2}{|S|^2 + 3\,|T|^2} \quad (8)$$

From a determination of the spin-averaged cross section and A_{ex}, one can determine both $|S|$ and $|T|$.

Determination of the phase difference between S and T requires, again in analogy with the discussion of Sec. 2, interference between the singlet and triplet channels. For example, in the antiparallel configuration of the scattering scenarios in Eqs. 4 and 5, the incident spin state is a linear superposition of singlet and triplet. The "interference" determines the relative cross section for reversing the incident electron's spin. Unfortunately, such a measurement would suffice to determine only the magnitude of the phase difference, not its sign. If, however, the incident electron's polarization were orthogonal to that of the incident atom, then the phase difference between S and T could be determined from the electron spin polarization direction after scattering.

Studies of spin dependence due to exchange are not yet as mature as studies of the spin-orbit interaction and there are to date no reports of a complete determination of the two scattering amplitudes. There are, however, several groups which have made partial measurements, from the pioneering work of Collins, et al.[25] and Hils, *et al.*.[26] on scattering from potassium, and Jaduszliwer, *et al.* on scattering from rubidium,[27] to the determined efforts of the Yale and City College groups to study scattering from atomic hydrogen,[28] to the work in Bielefeld on scattering from lithium[29] and our work at NIST on scattering from sodium.[30]

These experiments represent a rather wide range of interesting experimental approaches, and it is unfortunate that space constraints prevent describing each in detail. We show here as examples the results from two studies of the elastic scattering of spin-polarized electrons from spin-polarized alkali atoms. First we describe measurements made in Bielefeld of elastic scattering from polarized lithium atoms.

Spin-polarized electrons were scattered from a beam of atomic lithium which was spin-polarized in a hexapole magnet. The direction of atomic polarization was determined by diabatic or adiabatic passage through a magnetic field whose direction reverses over a short flight path of the atoms. The spin of neither the scattered elec-

[‡]We follow the unfortunate historical convention of referring to the two scattering amplitudes relevant for both the spin-orbit interaction and exchange by the same pair of symbols, f and g.

tron nor scattered atom was determined. The scattering asymmetry defined above for exchange, A_{ex}, was measured at fixed angles as a function of incident electron energy. These results are shown in Fig. 2. Also shown are the theoretical results of close-coupling[31,32] and modified polarized-orbital calculations.[33] The close-coupling results are in substantially better agreement with the data.

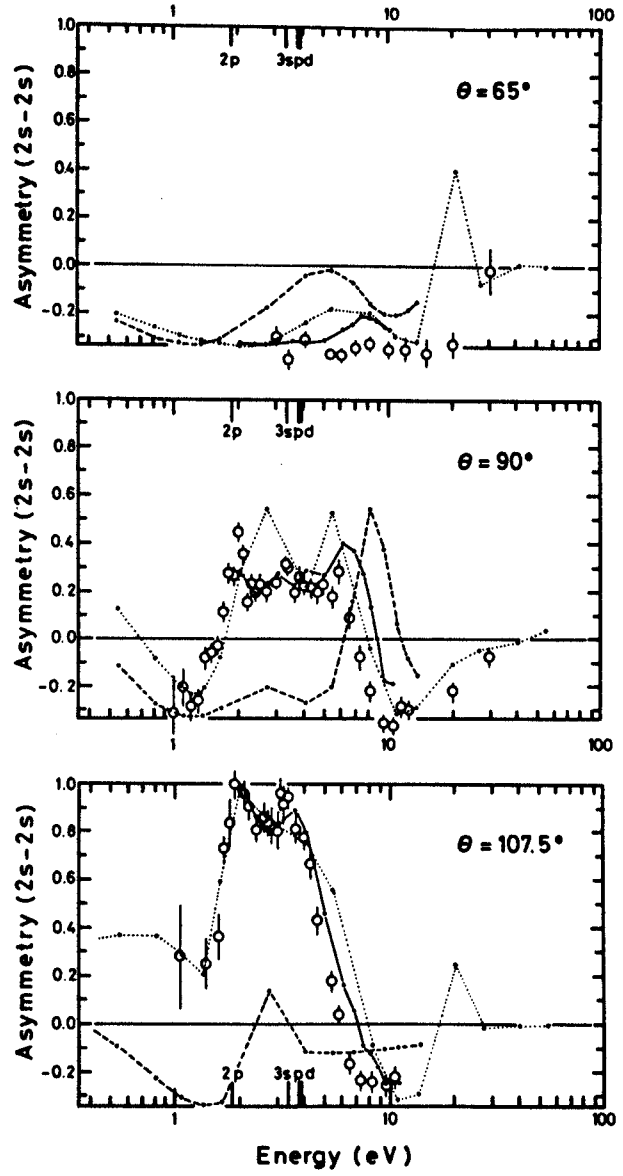

Fig. 2. Elastic spin asymmetry for scattering from Li. Experimental data from Baum *et al.*[29] Theoretical curves from Moores[32](—), Burke and Taylor[31](· · · ·), and Bhatia *et al.*[33](- - - -).

Two general features of the scattering are worth noting. First, for lower energies, below and slightly above the ionization threshold, this exchange asymmetry is very large. Limiting asymmetry values of $+1$ and $-\frac{1}{3}$ indicate pure singlet and triplet scattering, respectively. The scattering at 65° is dominated by triplet scattering, while the scattering to 107.5° is strongly dominated by singlet scattering. The second interesting general feature is that exchange continues to play an important role to relatively high incident energy, several times the ionization threshold.

We have made similar measurements at NIST for elastic scattering from sodium. Laser optical pumping was used to prepare ground state atoms polarized normal to a scattering plane. Electrons, also spin polarized normal to the plane, were scattered into angles in the range from 20° to 135°. As in the Bielefeld measurements, we measured the asymmetry A_{ex} for scattering with incident anti-parallel versus incident parallel spins. In Fig. 3 are shown results of our measurements for an incident energy of 54.4 eV. Even at this relatively high energy, about ten times the ionization threshold, exchange is seen to play an important role in the scattering.

Also included in Fig. 3 are results of a two-state close-coupling calculation of Mitroy, *et al.*[34] A four-state close-coupling calculation of Oza[35] gives near identical theoretical results. Though there are marked differences between the theory and experiment, there is agreement in a qualitative sense. The theory predicts correctly the size of the slight predominance of triplet scattering in the near forward direction, albeit not in the correct angular range. The source of this discrepancy is at present not understood.

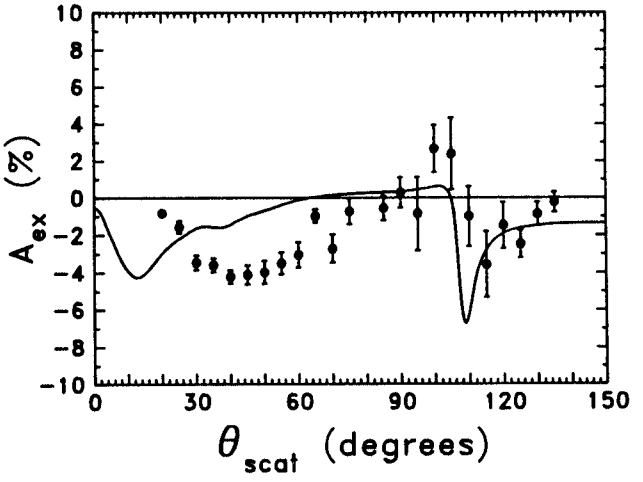

Fig. 3. Elastic spin asymmetry for scattering from Na at 54.4 eV. Experimental data from McClelland *et al.*[30] Theoretical curve from close-coupling calculation by Mitroy, *et al.*.[34]

One further interesting note from these elastic scattering measurements on Li and Na is that, contrary to conventional wisdom "exchange" is not necessarily most

important at larger scattering angles. For example in sodium at an incident energy of 54.4 eV, we find that exchange effects are most apparent for scattering angles between 30° and 60°.[30]

In our discussion thus far of exchange in elastic scattering from the alkalis, we have specifically neglected two important effects. We have neglected any effect of the spin-orbit interaction on the scattering and we have not considered exchange in scattering from targets with no net spin.

3.3 Exchange and Spin-Orbit Scattering

For the case where both the spin-orbit interaction and exchange are important, the formalism is substantially more complicated, with a total of six complex amplitudes required rather than the two complex amplitudes required for a description of either exchange or the spin-orbit interaction alone.[36] Though it is possible to devise a set of eighteen measurements which would be sufficient to determine this full set of amplitudes,[37] there are to date no reports of such a complete measurement.

One interesting feature of the simultaneous working of exchange and the spin-orbit interaction is a cooperative effect which should occur if both are present, but must vanish in the absence of either.[38,39] One should look for a left/right scattering asymmetry in the elastic scattering of unpolarized electrons from atoms polarized normal to the scattering plane. Equivalently, one would look for an asymmetry in the scattering of unpolarized electrons from "up" and "down" atoms. Neither the spin-orbit effect nor exchange could alone generate such an asymmetry.

In our measurements of spin dependence in elastic scattering from sodium at 54.4 eV, we have observed both an exchange and a spin-orbit asymmetry near the cross section minimum at about 110° scattering angle.[30] From those measurements we were also able to determine this joint asymmetry. However, within our present experimental uncertainty, we have not yet found evidence for this interesting cooperative effect.

3.4 Singlet → Triplet Excitation

The other important effect ignored in the above simplified description of exchange in electron–atom scattering is exchange with a target which has closed electronic shells and hence no net spin. In collision with such a target, an exchange event which changes the spin of the electron must also change the spin of the atom. As a result of the Pauli exclusion principle, this is not possible for elastic scattering. This is not to say that exchange with the spin-zero core of an alkali or with any spin-zero atom is not important — exchange certainly does contribute to the scattering cross-section and must be treated correctly before agreement with experiments can be reached. It does say that observable spin dependence in scattering from spinless atoms can appear only in conjunction with excitation to higher electronic states.

For example, excitation of triplet P levels in helium can proceed only via exchange and provides a unique opportunity to study exchange in electron atom collisions without use of spin-polarization techniques. While triplet excitation provides a direct measurement of an "exchange" process, there is no simple comparison with the analogous "direct" process, which would result in excitation of a singlet state in helium. Because of the energy difference between the singlet and triplet levels, far more than exchange determines the difference between singlet and triplet excitation amplitudes. Nevertheless, the results of such measurements contribute greatly to our overall understanding.

Excitation of the He(3^3P) level has been studied by groups in Stirling,[40] Belfast,[41] Utrecht,[42] and Perth.[43] We use the work of the Stirling group as an example. Electrons which excite ground state helium to the 3^3P state were detected in coincidence with the photon subsequently emitted from the excited state. The circular polarization of photons emitted normal to the scattering plane and linear polarization of photons emitted parallel to the scattering plane was determined. From these polarized coincidence measurements the three parameters λ, χ, and γ, which completely characterize the scattering process, could be determined.[5] Results of the measurement are shown in Fig. 4. Also shown are results of a first-order many-body theory calculation.[44] The general agreement of the model calculation with the experimental results is encouraging, but because of the extreme difficulty of the measurements and the consequently large experimental uncertainties, one cannot yet draw firm conclusions about agreement or disagreement between the theory and experiment.

4 Spin Dependence in Inelastic Electron Scattering

Having set the stage in the separate discussions of excitation and spin-dependent scattering, we now generalize the ideas of the previous sections and discuss the spin dependence observed in orientation and alignment studies of atomic excitation. As in the case of elastic scattering, we discuss separately those scattering processes in which the spin-orbit interaction and exchange play the central role.

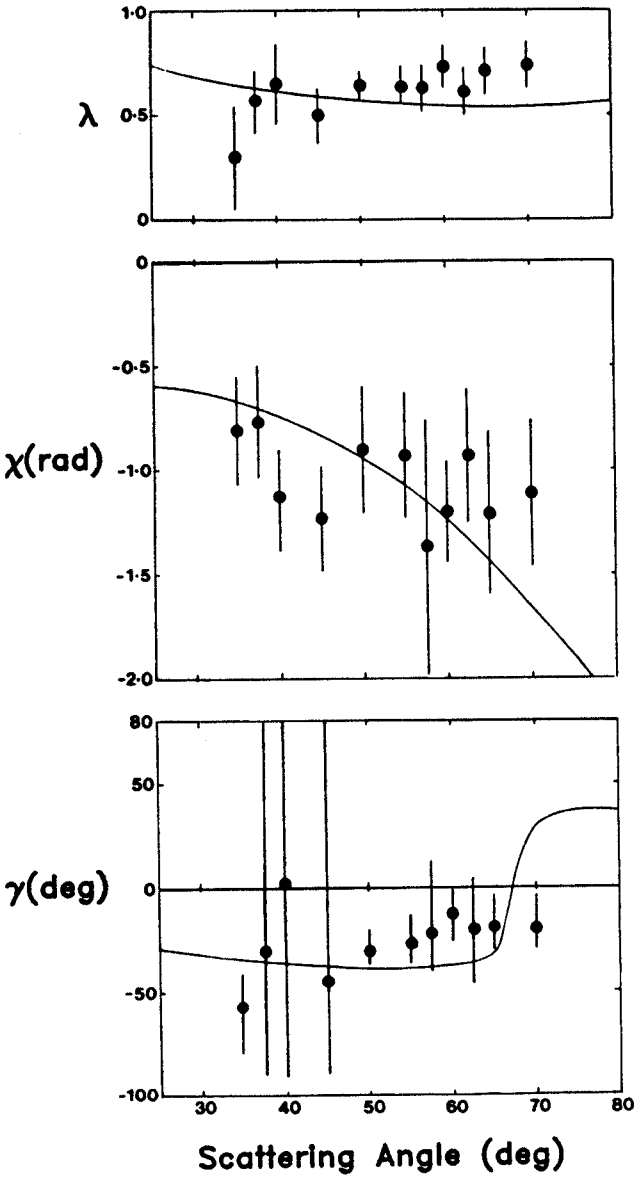

Fig. 4. The scattering parameters λ, χ, and γ from the Stirling measurements of He(3^3P) excitation at 60 eV.[40] Theoretical curve from FOMBT calculation of Cartwright and Csanak.[44]

4.1 Excitation of Hg(6^3P_1)

Electron impact excitation of heavy atoms is a good testing ground for state-of-the-art calculations of electron scattering. One must contend with the simultaneous importance of both exchange and the spin-orbit interaction, with transitions between multiple magnetic sublevels, with a rather complex atomic structure for which *LS*-coupling is a poor approximation, and with an often rich resonance structure for the colliding system.

Because all of these aspects of the collision interact in a complicated way, they can be very difficult to disentangle in order to extract an intuitive understanding of the collisional process. Methods of spin polarization and state selection can prove extremely valuable aids in attempts to unravel the details of this complicated collision system.

We take as an example the work of the Münster group for excitation of the 6^3P_1 level in mercury.[45] A detailed theoretical analysis of this system is too lengthy for the present, except to mention that a total of six complex amplitudes are required for a complete characterization of this collision system. While current experimental techniques do not allow a complete determination of this full set of amplitudes, the results of this measurement provide one of the most detailed characterizations yet of any collisional process.

Electrons, transversely polarized normal to the scattering plane, excited the target atoms and were detected in coincidence with fluorescence photons subsequently emitted perpendicular to the incident electron's direction, both in and perpendicular to the scattering plane. Excitation of the different magnetic sublevels, assumed quantized in the "collision" frame, was distinguished by the linear polarization of the photons. Photons from the $M_L = 0$ level are polarized parallel to the quantization axis while those from the $M_L = \pm 1$ are polarized perpendicular to it.

From the coincidence signals were derived six spin asymmetries. For each magnetic sublevel were determined:

$S_A(M_J)$ – the left/right scattering asymmetry for excitation by an initially polarized electron, averaged over the emission angle of the photon

$S_P(M_J)$ – the spin polarization after scattering of initially unpolarized electrons, also averaged over the emission angle of the photon.

The corresponding asymmetries, S_A and S_P, averaged over excitation of the different magnetic sublevels, were also determined.

Shown in Fig. 5 are the measurement results for these six asymmetries. Also shown in Fig. 5 are results of an *R*-matrix calculation from Bartschat *et al.*[46] The overall good quality of the agreement between theory and experiment is testimony to the current state-of-the-art for such scattering calculations.

The work of the Münster group also included similar studies of 6^3P_1 excitation at an incident energy of 15 eV. At the higher energy, the theory is not yet capable of accurately modeling the collision data.

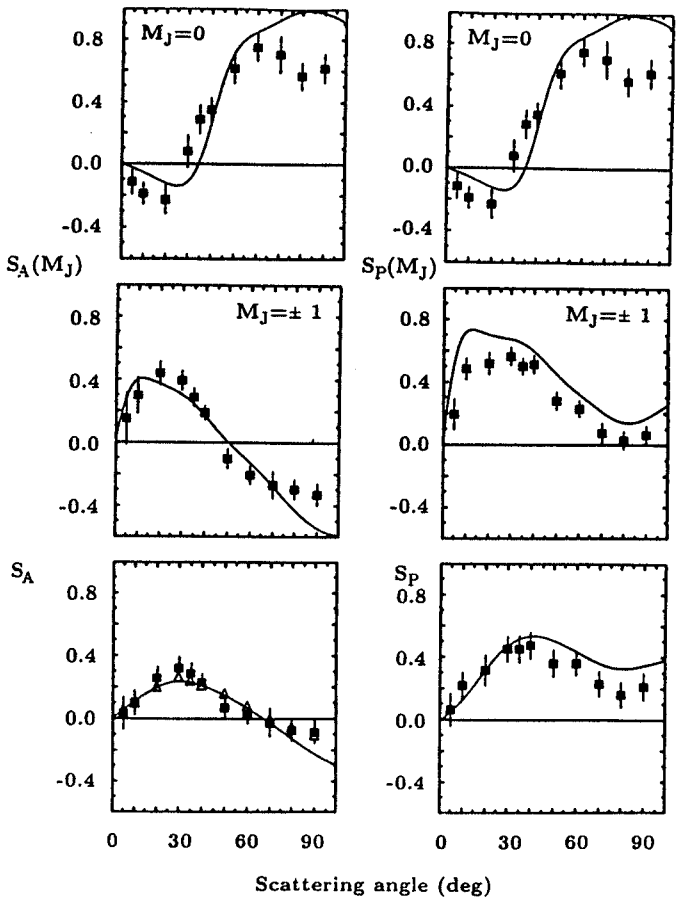

Fig. 5. Spin asymmetry functions $S_A(M_J)$, $S_P(M_J)$, S_A, and S_P at incident energy of 8 eV. Experimental results from Goeke, *et al.*[45] Theoretical curve from the R-matrix calculation of Bartschat, *et al.*[46]

4.2 Spin Dependence in Na 3P

We take as our final topic the spin dependence in $S \rightarrow P$ excitation of one-electron atoms. As noted in Sec. 3.2, electron scattering from alkali atoms provides a particularly good system for the study of exchange because an observation of the dependence of some scattering process on the relative spin orientations of the incident particles provides a direct measure of the effect of exchange on that collision.

Because a thorough treatment of the theoretical details necessary for an accurate description of this scattering process are available elsewhere,[47,48] we provide here only a brief introduction to aid in understanding of the experimental results. We take as the starting point the simplified description of $S \rightarrow P$ excitation provided in Sec. 2. Two complex scattering amplitudes were required for that case, one for excitation of each of two allowable magnetic sublevels. But now, as in the case of elastic scattering, there is a different amplitude for each relative orientation of the incident spins, or equivalently, each of the two incident composite spin states, singlet and triplet. This gives a total of four complex amplitudes to be determined, $f^S_{\pm 1}$ and $f^T_{\pm 1}$.

It is also, of course, possible to describe this collision process in the "collision" frame and to use uncoupled wavefunctions for the spins of the electron and atom. In this case, one would use, perhaps, the amplitudes f^d_0, f^d_1, f^e_0, and f^e_1 for "direct" and "exchange" excitation of the $M_L = 0$ and $M_L = 1$ sublevels.

We will discuss two rather different approaches for the determination of the amplitudes for spin-polarized excitation in alkali atoms. The first is the method used at JILA by Han, *et al.*[49] to measure directly the excitation of the four M_J sublevels of the $3P_{\frac{3}{2}}$ excited state in sodium, starting from the $M_J = +\frac{1}{2}$ ground state. Excitation of these four sublevels corresponds rather closely to collision events in which the orbital angular momentum M_L either does or does not change by 1 during collisions in which the electrons either do or do not "exchange". Han *et al.* denote the partial cross sections for these events by $Q^{|\Delta m_S|}_{|\Delta M_L|}$, with $|\Delta m_S|$, $|\Delta M_L| = 0$ or 1. Here $|\Delta M_L| = 0$ and 1 refer to excitation of the $M_L = 0$ and 1 sublevels in the collision frame. $|\Delta m_S| = 1$ and 0 refer respectively to changing or not changing the atomic spin in the collision.

The experimental technique was based on optical pumping to prepare a pure $M_J = +\frac{1}{2}$ initial state, and a spectroscopic determination of the M_J level excited by electron impact. A magnetic field of about 220 Gauss was applied to the collision region to split the magnetic sublevels of the excited state so that they could be resolved spectroscopically. This magnetic field had no fundamental effect on the electron impact excitation process.

The relevant energy level diagram is shown in Fig. 6. Laser optical pumping prepares spin polarized atoms in the upper of the two ground state sublevels. Electrons incident along the direction of the applied magnetic field collisionally excite the polarized atoms and the populations in the four $3^2P_{\frac{3}{2}}$ magnetic sublevels are determined by tuning a laser through the $3P_{\frac{3}{2}}(M_J) \rightarrow 5S_{\frac{1}{2}}(m_S)$ transition manifold and detecting the $4P \rightarrow 3S$ cascade fluorescence from the $5S$ state.

From the observed sublevel populations, Han *et al.* were able to extract the four partial cross sections mentioned above. Measurement results for these partial cross sections are shown in Fig. 7. Also shown are the results of a four-state close-coupling calculation from Moores and Norcross[50] which are overall in good qualitative agreement. It is unfortunate that this useful technique has very limited angular resolution and lacks a generalization

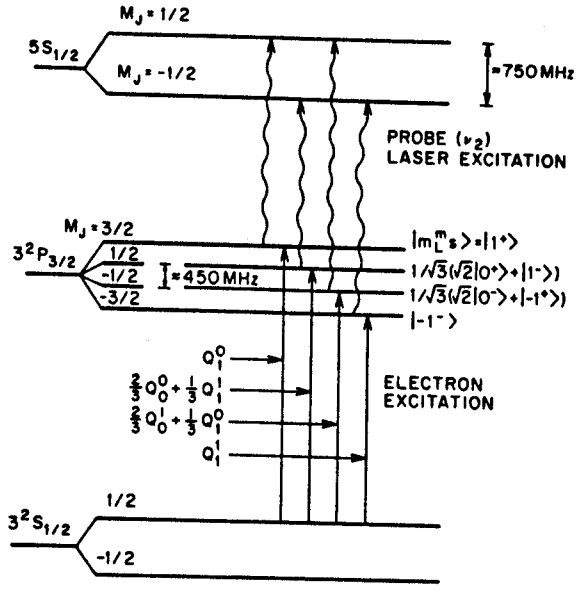

Fig. 6. Energy level diagram showing principle of technique used by Han *et al.*.[49]

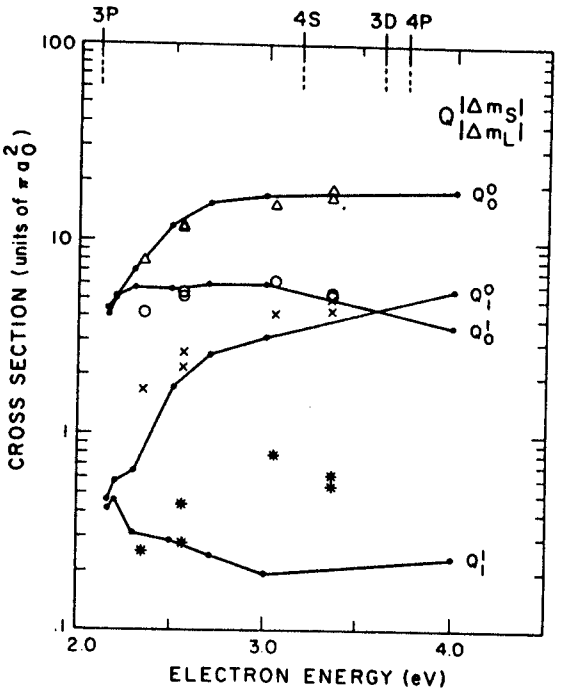

Fig. 7. Partial cross sections $Q^{|\Delta m_S|}_{|\Delta M_L|}$ measured by Han *et al.*,[49] compared to close-coupling calculation of Moores and Norcross.[50]

for determination of the phase relationship between the underlying scattering amplitudes.

Conventional crossed-beams experiments which include spin polarization of both the incident electrons and atoms in angle resolved collision studies have also been carried out. At NIST, we have studied this process using electron impact de-excitation of laser-excited sodium atoms. Similar measurements have been performed in Bielefeld using excitation of spin-polarized lithium atoms by spin-polarized electrons.[51]

The apparatus at NIST for these measurements is the same as is used for the elastic scattering measurements described in Sec. 3.2 except that the optical pumping laser illuminates the scattering volume directly to maintain a population of excited and optically pumped atoms. The primary advantages of optical pumping over magnetic state selection and electron-photon coincidence methods are that it allows studies of either elastic or inelastic transitions with the same apparatus, and that the inelastic measurements have a much larger scattering signal than the equivalent electron-photon coincident experiments.

At each incident energy and scattering angle, four count rates were recorded, corresponding to the four relative spin orientations of the incident electrons and atoms. From these four count rates were derived three relative quantities, the ratio between the triplet and singlet scattering cross sections, $R = \frac{|T|^2}{|S|^2}$, and angular momentum transferred normal to the scattering plane, separately for the singlet and triplet channels, $L_\perp^S$ and $L_\perp^T$. Because no spin analysis is performed after collision, no information is accessible about the phase differences between the scattering amplitudes, except for the spin-averaged phase difference determined from scattering from atoms excited with linearly polarized light as discussed in Sec. 2.

In Fig. 8 we show results of our measurements of these quantities at an incident energy of 2.0 eV. Also shown, in Fig. 8a, is the spin-averaged $L_\perp$ both from our measurements and from Hermann, *et al.*[52] The theoretical results of Moores and Norcross[50] are included in Fig. 8 for comparison. Several features are worth noting. First, the generally good agreement apparent in Fig. 8a for the unpolarized $L_\perp$ at smaller scattering angles is misleading. The substantial disagreement for the singlet channel angular momentum observed in Fig. 8b is masked in the unpolarized data by the much larger contribution of the triplet state to the unpolarized measurements due to its degeneracy factor of three. Because the triplet channel shows good agreement at small angles, the disagreement for singlet scattering is masked.

The second interesting feature is that while the theoretical results for the triplet channel are in better agree-

ment at smaller scattering angles, the situation is reversed at larger angles. At larger angles the calculation is better for the singlet channel and drastically overestimates the triplet channel angular momentum transfer for angles larger than about 90°. The marked discrepancy apparent in Fig. 8a for scattering larger than about 110° comes entirely from the triplet channel. Over the intermediate angular range from about 45° to 110° the discrepancy is shared more or less equally between the two channels.

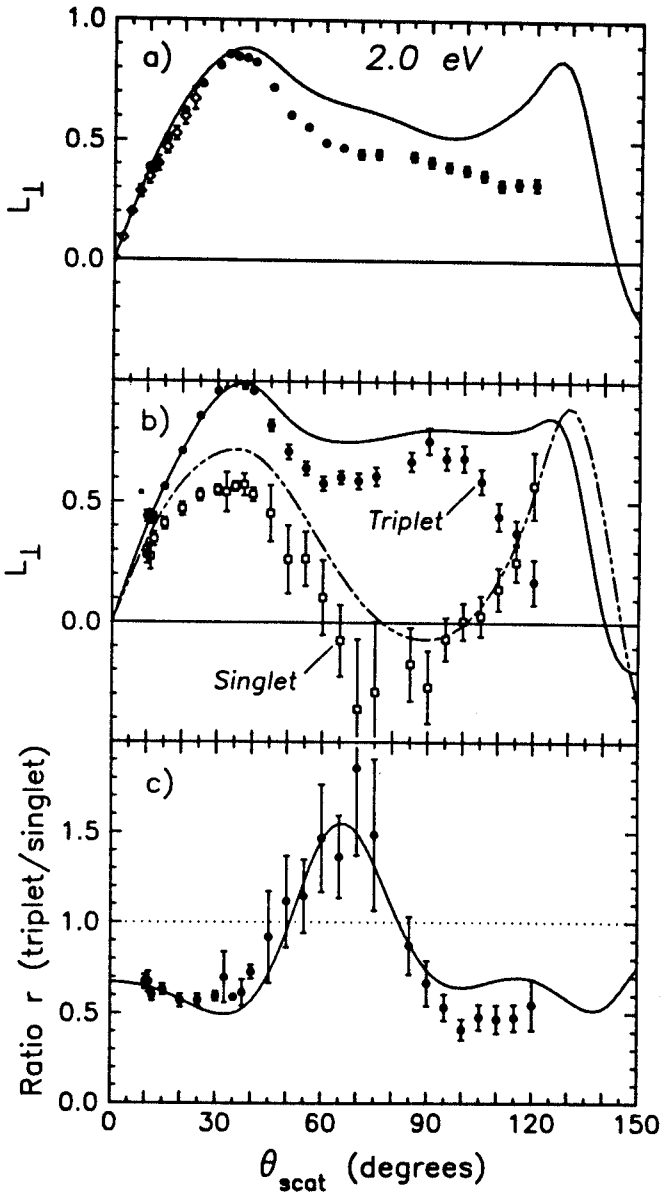

Fig. 8. $L_\perp$, $L_\perp^S$, $L_\perp^T$, and R measured at NIST for Na($3P \rightarrow 3S$) de-excitation at 2.0 eV.[53] Theoretical curves are close-coupling results from Moores and Norcross.[50]

Finally, we note that the calculation reproduces very well the ratio of triplet to singlet scattering show in Fig. 8c. Over much of the angular range, singlet scattering is dominant by roughly a factor of two. In a rather narrow range, from about 55° to 80°, triplet scattering becomes about 50% larger than singlet.

In the spirit of "complete" measurements, as put forward in Sec. 2 of this work, we have been attempting to make detailed measurements of both the elastic and the dominant inelastic channel for each incident energy. We have completed measurements of elastic and inelastic scattering at an incident energy of 54.4 eV ,[53–55] and are just completing a similar series of measurements at 20.0 eV.[56]

5 Summary

We have seen that great strides have been made toward a quantitative understanding of electron–atom collisions, and that state-selection techniques continue to play a very important role in the advancement of our knowledge. The extension of alignment and orientation studies to include spin dependence has brought us closer to realization of the goal of "complete" measurements for electron–atom collisions. Nevertheless, there is much work yet to be done before the experimental characterization of these collisions is complete.

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