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LETTER TO THE EDITOR

Spin-resolved superelastic electron scattering from laser-excited chromium atoms

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Abstract. Superelastic scattering of polarized electrons from laser-excited polarized chromium atoms ($4^7P^o_4 \rightarrow 4^7S_3$ transition, energy gain 2.92 eV) has been measured for total collision energies of 6.8 and 13.6 eV (0.5 and 1.0 Ryd) and scattering angles ranging from 10° to 140°. The orientation parameter $L_\perp$ is determined both averaged over spin states, and separately for the sextet and octet total spin channels. The octet-to-sextet cross section ratio r is also determined. The chromium atom has six unpaired electrons with the configuration $3d^5(^6S_{5/2})4l$ ($l = 0, 1$) for the states under investigation. The scattering data show strong orbital orientation and spin effects, which we attribute to the interaction between the continuum electron and the outermost $4l$ target electron.

In the study of electron–atom collisions, state selection has played a major role in helping to provide benchmark experimental results for evaluation of theoretical approaches. State selection of the electrons being scattered, through the use of spin-polarized electron sources and spin detectors, has made a major contribution to our understanding of electron–atom scattering processes by elucidating the roles played by both the spin–orbit and the exchange interactions (Kessler 1985). State selection of the target atoms, through laser optical pumping or electron–photon coincidence studies, has similarly led to new insights, particularly in the realm of angular momentum transfer during the collision (Andersen *et al* 1988). To fully elucidate the scattering process, the combined effects of angular momentum transfer and electron spin have been studied by using both spin-polarized electrons and state preparation of the atomic targets. Sodium has provided a good test case for this type of study when exchange is the dominant spin effect, because it is essentially a one-electron target with a simple S $\rightarrow$ P transition (McClelland *et al* 1989, Nickich *et al* 1990, Scholten *et al* 1991).

We present here measurements of superelastic scattering of spin-polarized electrons from optically pumped chromium atoms. Unlike electron–sodium scattering, the electron–chromium scattering system provides the opportunity to study exchange in a multi-electron system with high net spin. The chromium states involved in the present study are the septet levels 4^7S_3 (ground state) and $4^7P^o_4$. These states, with six valence electrons in the configuration $3d^54l$, where $l = 0$ or 1, have a net electronic spin S of 3; that is, all six valence electron spins are parallel.

Exchange in electron scattering from a high-spin target such as chromium has not been studied to date, either experimentally or theoretically. In considering such a system, a number of new issues arise with regard to the relative roles played by the valence electrons.

For example, in superelastic scattering, the extent to which the net spin of the the 3d-shell electrons is decoupled from the spin of the 4*l* electron during the collision is of interest. In other words, is there, for spin–spin coupling, an equivalent to the Percival–Seaton hypothesis (Percival and Seaton 1958), which states that orbital angular momentum can be decoupled from electron and nuclear spin during the collision?

Studying electron–chromium scattering is also motivated in a broader sense. There are a large number of atoms which have an open-shell and a high-spin configuration, yet very few have been the subject of much experimental investigation. These must be studied experimentally and understood theoretically to obtain a unified picture of electron–atom scattering.

Our experimental studies on chromium use essentially the same apparatus used for spin-polarized electron–sodium scattering studies (McClelland *et al* 1989). A spin-polarized electron beam, produced in a GaAs source (Pierce *et al* 1980), is incident upon a region of optically pumped atoms. The scattered electron intensity is measured as a function of incident energy, scattering angle, and the relative orientation of the electron and atom spins, which are perpendicular to the scattering plane. The electron beam had a spin polarization of $P_e = 0.33$, as measured by a cylindrical Mott polarimeter (Hodge *et al* 1979). The uncertainty in the polarization, dominated by systematics in the calibration of the polarimeter, is about ± 0.02. The electron beam energy was calibrated by introducing a background gas of helium and observing the resonance at 19.3 eV in elastic scattering at 90°. The energy spread, also measured by this method, was 0.2 ± 0.1 eV. The chromium atoms were produced in an oven with a tantalum crucible heated by electron bombardment to about 1400 °C. Skimmers and apertures were used to define a 4 mm diameter atomic beam at the scattering centre.

The $4^7S_3 \rightarrow 4^7P_4^o$ transition was optically pumped with circularly polarized light at a wavelength of 425.43 nm. Several hundred milliwatts of laser light at this wavelength were obtained from a UV-pumped single-frequency dye laser operating with stilbene 420 dye. A 1 cm length of the atomic beam was pumped, producing a population of spin-polarized excited-state chromium atoms suitable for performing superelastic scattering measurements. The polarization P_a of the excited chromium atoms, which is related to the relative populations of the magnetic sublevels M_J, was examined by observing the degree of linear polarization of the fluorescence light that was emitted in the scattering plane from the excited atoms at the target. These measurements indicated that P_a was essentially 1.0, with uncertainty less than typical measurement errors. A P_a of 1.0 corresponds to an excited state population that is concentrated entirely in the $M_J = +4$ (or -4) state.

Four different count rates were measured: $I_{\uparrow\uparrow}$, $I_{\uparrow\downarrow}$, $I_{\downarrow\uparrow}$, and $I_{\downarrow\downarrow}$, where the arrows denote the orientation of the atoms and the electron spin, respectively. Superelastically scattered electrons, with count rates at least 4 s^{-1}, were counted for up to 30 min at each scattering angle and incident energy, switching the electron spin at 100 Hz with a Pockels cell and changing the atom polarization from up to down by rotating a quarter-wave plate at intervals of up to 20 s. In addition, a background signal was measured by periodically blocking the laser beam. From the count rates with background subtracted, a series of partial intensities were calculated:

$$I_{tot} = I_{\uparrow\uparrow} + I_{\uparrow\downarrow} + I_{\downarrow\uparrow} + I_{\downarrow\downarrow} \tag{1}$$

$$O = (I_{\uparrow\uparrow} + I_{\uparrow\downarrow}) - (I_{\downarrow\downarrow} + I_{\downarrow\uparrow}) \tag{2}$$

$$S = (I_{\uparrow\uparrow} + I_{\downarrow\uparrow}) - (I_{\downarrow\downarrow} + I_{\uparrow\downarrow}) \tag{3}$$

$$A = (I_{\uparrow\downarrow} + I_{\downarrow\uparrow}) - (I_{\uparrow\uparrow} + I_{\downarrow\downarrow}). \tag{4}$$

These partial intensities were combined to generate the observables of the experiment.

The observables we present are chosen to highlight angular momentum transfer and exchange in the collision. The quantity $L_\perp$ represents the angular momentum transferred perpendicular to the scattering plane in the collision. Because we are using spin-polarized electrons, $L_\perp$ can be resolved into the contributions of the individual spin channels of the scattering process. When the spin of the continuum electron is coupled to the spins of the valence electrons, the resulting net spin is either $S = \frac{7}{2}$ (octet) or $S = \frac{5}{2}$ (sextet). If we assume that the spin–orbit interaction can be ignored because chromium is a relatively light target ($Z = 24$), it follows that the total spin is conserved and the sextet and octet spin states are not mixed in the collision process. The sextet and octet channels are then the most appropriate for describing the scattering. We determine

$$L_\perp = \frac{1}{P_a}\frac{O}{I_{tot}} \tag{5}$$

$$L_\perp^s = \frac{P_e O - \frac{4}{3}P_a S}{P_e P_a I_{tot} - \frac{4}{3}A} \tag{6}$$

$$L_\perp^o = \frac{P_e O + P_a S}{P_e P_a I_{tot} + A} \tag{7}$$

$$r = \frac{3P_e P_a I_{tot} - 3A}{3P_e P_a I_{tot} + 4A} \tag{8}$$

where $L_\perp$ is the spin-averaged angular momentum transferred perpendicular to the scattering plane, $L_\perp^s$ and $L_\perp^o$ are the values of $L_\perp$ for the sextet and octet channels, and r is the ratio of octet to sextet cross sections, averaged over angular momentum transfer channels.

The spin-averaged orbital orientation parameter $L_\perp$ is non-zero because optically pumped excited chromium atoms are in pure orbital angular momentum states with magnetic quantum numbers $M_L = +1$ or -1, and the scattering depends on that orientation of the target (Andersen *et al* 1988). The sextet and octet versions of $L_\perp$ differ because exchange has an influence on this orientation dependence. The octet to sextet ratio r reflects the relative behaviour of the sextet and octet scattering channels averaged over the angular momentum transfer channels.

Our results are shown in figures 1 and 2 for total energy $E_{tot} = 13.6$ eV (1.0 Ryd) and 6.8 eV (0.5 Ryd) respectively. The total energy E_{tot} is the energy of the electrons *after* the superelastic collision, during which they gain 2.92 eV, so these total energies correspond to incident energies of 10.68 eV and 3.88 eV, respectively. The error bars shown in the figures represent the statistical uncertainties and do not include the systematic uncertainties of the polarizations.

At $E_{tot} = 13.6$ eV, the measured scattering angles range from 10° to 40°. We observe a large spin-averaged $L_\perp$ at this energy, as shown in figure 1(*a*). The behaviour is quite similar to that observed in scattering from other atomic targets with S $\rightarrow$ P transitions (Andersen *et al* 1988). $L_\perp$ is positive and increasing for small scattering angles, then peaks around 25° and decreases toward zero at about 40°. Figure 1(*b*), which shows $L_\perp^s$ and $L_\perp^o$, indicates a significant difference between these two parameters. As in the case of sodium (McClelland *et al* 1989, Scholten *et al* 1991), the lower net spin channel appears to have a smaller value of $L_\perp$ at these small angles. The ratio r is shown in figure 1(*c*). Surprisingly, the ratio is essentially 1.0 throughout most of the angular range. This should not be interpreted as a lack of exchange effects, however, because of the significant difference between $L_\perp^s$ and $L_\perp^o$ seen in figure 1(*b*). These three parameters are completely independent, and the

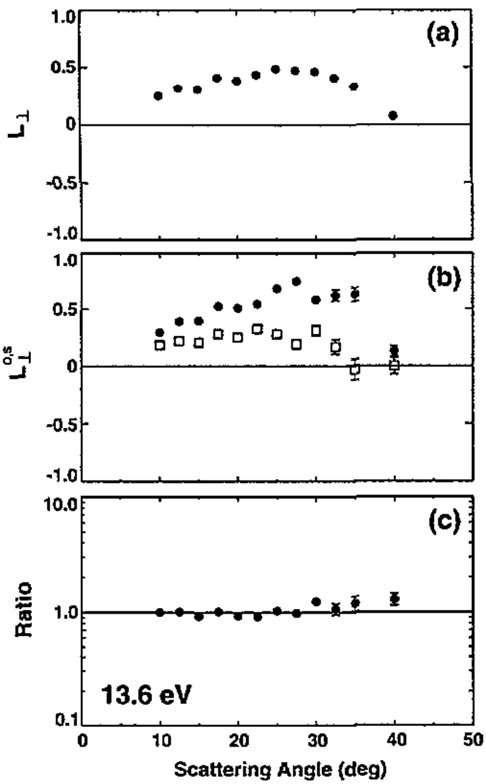

Figure 1. Superelastic electron–chromium scattering with $E_{tot} = 13.6$ eV total collision energy, plotted against scattering angle. (*a*) Spin-averaged angular momentum transfer, $L_\perp$ (in units of $\hbar$). (*b*) Spin-resolved angular momentum transfer, $L_\perp^s$ (□) and $L_\perp^o$ (●). (*c*) Octet-to-sextet ratio, r. Uncertainty estimates are one standard deviation from counting statistics, and are shown only when larger than the plotting symbol.

behaviour of all three must be considered before drawing conclusions about the role played by exchange.

For the lower total scattering energy of 6.8 eV the angular range covered was much greater, extending to 140°. The signal-to-noise ratio of the experiment was better for the larger angles at this energy because the scattering cross section drops off less quickly than it does at 13.6 eV. Figure 2(*a*) shows the spin-averaged $L_\perp$, which again shows essentially the expected S → P behaviour, with the possible exception that no large negative dip is seen in the region from 50° to 100°. The sextet and octet $L_\perp$ values, shown in figure 2(*b*), indicate large exchange effects at this scattering energy, especially in the region from 20° to 40°, where $L_\perp^o$ reaches nearly 1.0 and $L_\perp^s$ only about 0.5. The octet-to-sextet ratio r is shown in figure 2(*c*). The ratio is below one for the entire angular range, indicating the dominance of the sextet channel. At this energy, exchange appears to influence r as well as $L_\perp^s$ and $L_\perp^o$.

The observations of spin-polarized superelastic electron scattering from chromium presented in this letter represent the first measurements of spin-polarized electron scattering from a high-spin, open-shell target. While theoretical calculations must be performed to determine this definitively, the fact that the angular momentum transfers and spin channel ratios behave qualitatively as they do in electron–sodium scattering suggests that the process

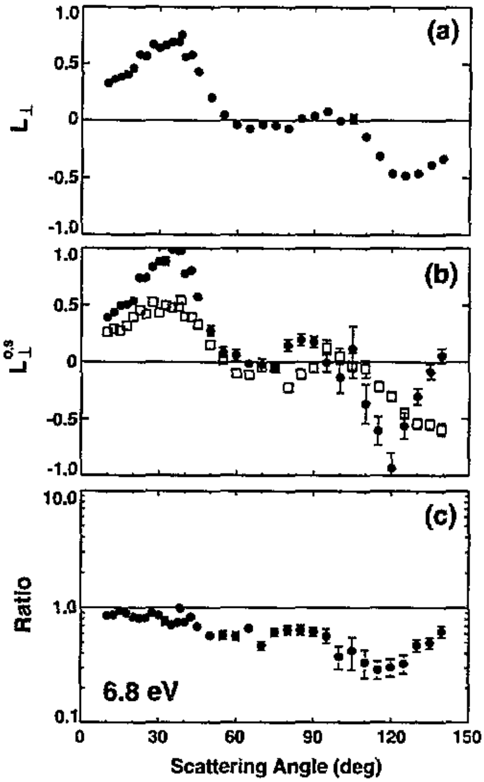

Figure 2. Superelastic electron–chromium scattering with $E_{tot} = 6.8$ eV total collision energy, plotted against scattering angle. (*a*) Spin-averaged angular momentum transfer, $L_\perp$ (in units of $\hbar$). (*b*) Spin-resolved angular momentum transfer, $L_\perp^s$ (□) and $L_\perp^o$ (●). (*c*) Octet-to-sextet ratio, r. Uncertainty estimates are one standard deviation from counting statistics, and are shown only when larger than the plotting symbol.

is essentially alkali-like. There is no evidence in the present data to suggest that the 3d electrons in the chromium atom play anything other than a spectator role in the superelastic scattering process. It is our hope that this work will stimulate theoretical investigations of the electron–chromium scattering system, so that some of the issues associated with multi-electron, open-shell atoms can be resolved. In addition, we hope that further experimental results will be forthcoming, especially on elastic scattering, where it is expected that the multi-electron nature of the atoms will play a much more important role.

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