Superelastic electron scattering on lithium

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Reduced Stokes parameters of the $2\,^2P$ state of lithium are measured using superelastic electron-scattering techniques and calculated using the convergent close-coupling method. The measurements and calculations are in excellent agreement at all scattering angles. This is particularly significant at the large scattering angles where there has been a long-standing discrepancy between theory and electron-photon coincidence measurements in the corresponding electron-hydrogen scattering problem. [S1050-2947(96)50207-6]

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There has been a very long-standing discrepancy at large scattering angles between theory and measurements of the $2\,^2P$ angular correlation parameters for 54.4-eV electron-impact excitation of atomic hydrogen. In our view, this is the most outstanding problem in fundamental electron-atom scattering. The development of most electron-atom scattering theories uses the e-H system as a testing ground for dealing with the more sophisticated problems. The discrepancy with experiment here undermines the basic building blocks of such theories. For this reason it is imperative that this problem be resolved as soon as possible.

The aim of this Rapid Communication is to address these issues. There have been two independent measurements [1,2] of the e-H $2p$ angular correlation parameters that are in agreement with each other. Both of these used the electron-photon coincidence technique where Lyman-α photons are detected in coincidence with electrons that have lost 10.2 eV of the initial 54.4-eV energy. These measurements are particularly difficult at the backward angles, as there the cross section is very small. The 54.4-eV energy is a particularly good choice because at this energy the total ionization cross section peaks and is of similar value to the integrated $2\,^2P$ cross section. Since the electron flux is evenly divided between the ionization and the $2\,^2P$ channels one would expect a theory to get both of these right before it could be confident of providing more detailed differential information.

The treatment of the ionization channels is one of the most difficult aspects in electron-atom scattering theory, and it was not until the introduction of the convergent close-coupling (CCC) theory that a single theory predicted the correct total cross sections for both $2\,^2P$ excitation [3] and ionization [4]. Nevertheless, this theory yielded results for the angular correlation parameters that were much the same as the previous most sophisticated theories. The CCC theory claims to treat both the discrete and continuum target subspaces to a demonstrated level of convergence, and simply relies on nonrelativistic quantum mechanics for governing the e-H scattering system. Thus we are left with the question: Why is there discrepancy between the CCC theory and the angular correlation experiments?

Madsen and Taulbjerg [5] argue that the special nature of the hydrogen atom, namely the degeneracy of the energy levels, will lead to a slow convergence with expansion in target-space angular momentum. Though the three CCC calculations that included s and p states, s, p, and d states, and s, p, d, and f states were demonstrated to vary by only a few percent [3], they suggested that even larger l states might be necessary. These conclusions were made on the basis of a distorted-wave eikonal theory that is unable to obtain agreement with experiment at the small scattering angles, where the CCC theory and many others have no difficulty. For this reason we suspect that the discrepancy is not due to convergence problems in the CCC theory.

An approach to this problem is needed from an experimental side. Another way to measure the angular correlations is by means of superelastic electron-scattering techniques. The target is prepared in the excited p state by means of a suitable laser. Unfortunately no such lasers exist to prepare the hydrogen atom in an initial $2p$ state. However, this can be done for the $2p$ state of lithium. As in hydrogen, the lithium total ionization cross section peaks at around 4 times the ionization threshold [6], and so we suggest that e-H scattering at 54.4 eV may be qualitatively compared with e-Li scattering at 20 eV.

The major components of the apparatus used in the present experiments have already been described in detail [7]. The experimental configuration modified for superelastic electron-scattering studies on lithium is shown in Fig. 1. Electrons at an energy of 20 eV are incident on ^{6}Li atoms in the excited state $2\,^2P_{3/2}$ prepared by optical pumping. The count rate of superelastic electrons scattered with energy 21.8 eV is measured as a function of scattering angle θ for different states of the laser polarization. From this one can define the reduced Stokes parameters:

$$P_1=\frac{1}{K}\frac{I_0-I_{90}}{I_0+I_{90}},\quad P_2=\frac{1}{K}\frac{I_{45}-I_{135}}{I_{45}+I_{135}},\quad P_3=\frac{1}{K'}\frac{I_{\sigma^-}-I_{\sigma^+}}{I_{\sigma^-}+I_{\sigma^+}},\tag{1}$$

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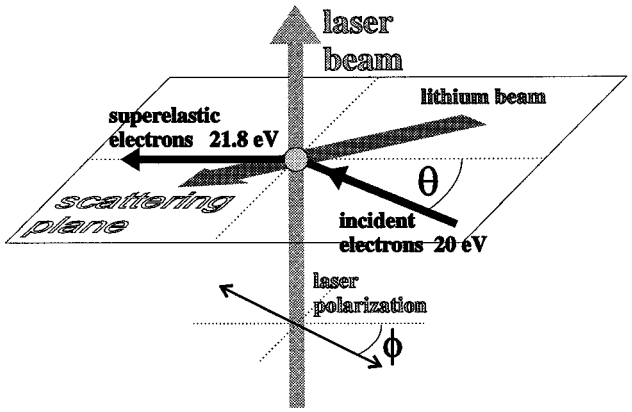

FIG. 1. A schematic diagram of the experimental geometry.

where I_n is the superelastic count rate when the laser radiation is polarized at n degrees to the outgoing electron direction, and $\sigma^{\pm}$ indicates left-hand $(+)$ and right-hand $(-)$ circular polarization. The line polarization P_L of the resonant fluorescence is also measured and taken to be identical to the superelastic depolarization factor K [8]. The factor K' is equal to the circular polarization of the resonant fluorescence emitted normal to the scattering plane and is taken to be unity [8].

The beam of lithium atoms was produced by a resistively heated oven. An estimated atom density at the interaction region was about 5×10^{10} atoms/cm^3 under normal operating conditions. The Doppler width of the beam was 50 MHz. Using a multistage electron gun with a barium oxide cathode, an electron beam at 20 eV was produced with an energy spread of 0.3 eV and typical current of 1–2 μA. The measured divergence of the electron beam was around 2°–3° (full width at half maximum).

Two types of electron-energy analyzers were used to measure the count rate of the superelastically scattered electrons. A cylindrical mirror analyzer was used for angles less than 15°, while a more sensitive retarding field analyzer was employed at the larger scattering angles. The overall energy resolution of the system was less than 0.5 eV, which was sufficient to separate the superelastic signal and the background of elastically scattered electrons. A typical count rate of superelastic electrons was around 20–40 Hz at the scattering angle of 90°. This angle corresponds to the minimum in the differential cross section for the $2S$-$2P$ transition.

Laser radiation at 670.977 nm was produced by a single-frequency stabilized ring dye laser. Typical laser power was 300 mW, corresponding to a laser intensity of 20 mW/mm^2 at the interaction region. In order to avoid radiation trapping and increase the excited-state fraction of lithium atoms in the interaction region it is necessary to pump both ground-state hyperfine levels simultaneously. A $LiTaO_3$ electro-optic phase modulator was used to generate a number of frequency-shifted sidebands. Two of them, separated by 228 MHz, were used to pump the ground-state levels $2\,^2S_{1/2}, F=1/2$ and $2\,^2S_{1/2}, F=3/2$ simultaneously. Using this technique, the fraction of the excited ^{6}Li atoms and consequently the superelastic scattering count rate has been increased by a factor of 2 when compared with single-frequency pumping.

Two quarter-wave plates mounted in mechanical rotators were used to obtain either circular polarization or any orientation of linear polarization of the laser radiation. Computer controlled stepper motors were used to rotate the waveplates. In this way all three Stokes parameters were measured during the same run.

A correct measurement of the superelastic depolarization factor K is essential for interpretation of the experimental results. To obtain this information the polarization of the decay fluorescence was monitored through the vacuum window and the factor $K=P_L$ was determined for each measurement. Measurements and calculations show that for the ^{6}Li transition $2\,^2S_{1/2}-2\,^2P_{3/2}$ the factor K is essentially independent of laser intensity during typical experimental conditions. The measured superelastic depolarization factor K was 0.570 ± 0.005.

In Fig. 2 we give the results of the measurements and the CCC calculation. The values of P_1, P_2, and P_3 were measured several times at both positive and negative scattering angles. The final values of each parameter have been determined by taking the weighted means of the several sets of data. The error bars represent plus or minus one standard deviation. These errors include statistical uncertainties and residual systematic effects, which have been identified as arising from the divergence of the electron beam, small dichroic effects introduced by the vacuum window and other components in the laser beam line, and slight misalignment of the electron beam. An estimate of the angular errors was obtained by measuring all three Stokes parameters at the angular ranges of $-100°$ to $-6°$ and 6° to 100°. This showed that the uncertainty of measurement of the scattering angle was less than $\pm1.5°$ for the forward angles, rising to $\pm2.5°$ for the backward angles.

The CCC theory for hydrogenic targets [9] was used, and convergence was found using $8s$, $9p$, $7d$, and $5f$ states. The incident energy was taken to be 21.8 eV so that the outgoing energy was 20 eV, the time-reversed situation of the experiment. We see that we have essentially quantitative agreement for all of the presented parameters at all measured scattering angles.

The Stokes parameters are trivially related to the λ, R, and I angular correlation parameters traditionally presented for hydrogen by

$$P_1=2\lambda-1,\quad P_2=-2\sqrt{2}R,\quad P_3=2\sqrt{2}I,$$

$$P^+=\sqrt{P_1^2+P_2^2+P_3^2}. \tag{2}$$

For comparison, we give the e-H case in Fig. 3. We see that the CCC theory predicts qualitatively similar behavior of the Stokes parameters for the two targets. There is a difference between the P_2 parameters at the small angles for hydrogen and lithium, where theory and experiment predict more structure in the lithium case. The deep minumum at the backward angles of P_2 is predicted by the CCC theory for both targets, but agreement is found only with the present measurements in lithium. Similarly, the agreement of the CCC

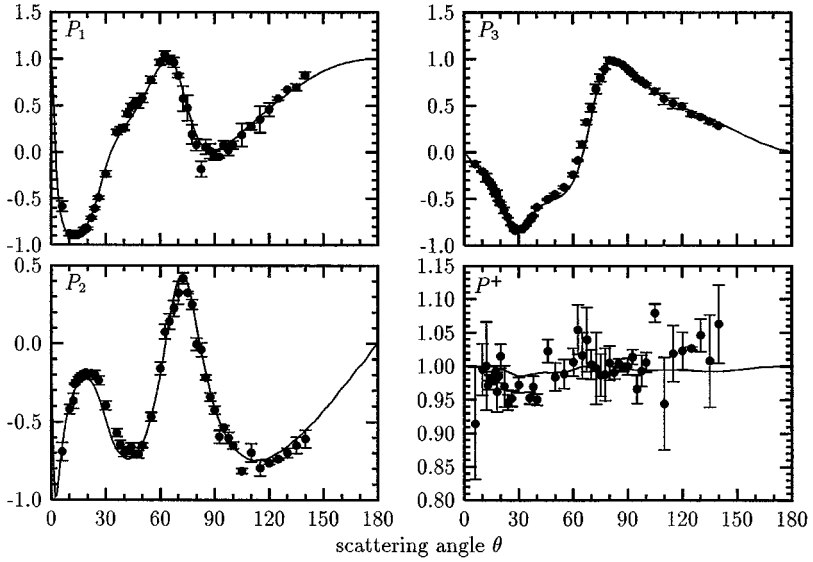

FIG. 2. Present measurements and calculations of electron-lithium $2p$ Stokes parameters at 21.8 eV. The measurements were obtained using superelastic techniques. The calculations were performed using the convergent close-coupling (CCC) theory.

theory with the present measurements at the deep minimum of the P_1 parameter at the intermediate angles is most encouraging, as this is not the case for hydrogen. Furthermore, agreement between theory and measurements of P_3 is excellent in the case of lithium, but not so in the case of hydrogen. For both systems the theory predicts $P^+ \approx 1$ over the entire angular range, though exchange scattering at these energies is quite substantial. The present measurements support this, but those for the e-H system do not. This parameter is not particularly useful for studying effects of exchange in substantial detail. To do this we need to look at the spin asymmetry; see Ref. [9], for example, in the case of e-Na scattering.

In conclusion, we have seen that there is quantitative agreement between the CCC theory and experiment for electron-impact excitation of the $2p$ state of lithium. The impact energy was chosen to allow an appropriate comparison with similar e-H measurements, which used an alternative experimental technique, and where there is substantial discrepancy with theory. These results give us greater confidence in the validity of the CCC and other sophisticated theories for the e-H problem, though direct verification is still warranted. We shall now perform measurements and calculations at a number of other energies for the e-Li system to ensure that the CCC theory is valid over the entire energy range, as is supposed.

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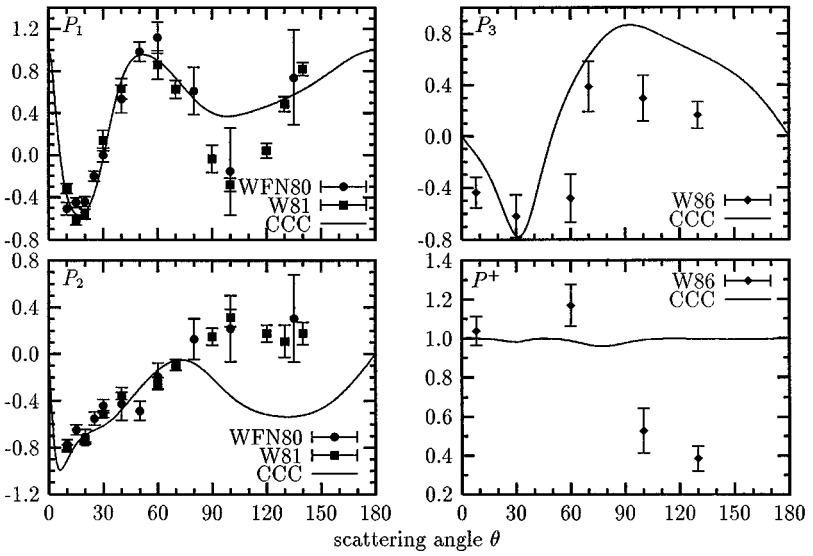

FIG. 3. Stokes parameters for electron-hydrogen $2p$ excitation at 54.4 eV. The measurements denoted by WFN80, W81, W86 are from Refs. [1,2,10], respectively. The theory is the 36-state CCC calculation [3].

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